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- Hydrazinecarboxamide derivatives, a process for production thereof, and uses thereof.
- The present invention relates to a hydrazinecarboxamide derivative of the general formula (I) shown below:

wherein the substituents are as defined in the specification, which has a wide insecticidal spectrum at a low dosage, a process for producing said derivative, and utilization of said derivative as an insecticide.

BACKGROUND OF THE INVENTION

Field of the invention

This invention relates to hydrazinecarboxamide derivatives, in particular, compounds having a wide insecticidal spectrum.

Related Art

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Jap. Pat. Appln. Kokai (Laid-Open) No. 48-91223 discloses that semicarbazides are effective as insecticides. Jap. Pat. Appln. Kokai (Laid-Open) No. 54-119029 discloses that thiosemicarbazones are useful as agricultural and horticultural disease-controlling agents. Jap. Pat. Appln. Kokai (Laid-open) No. 63-93761 discloses that substituted hydrazones are effective as pest-controlling agents.

However, a compound having a wide insecticidal spectrum has not yet been disclosed.

SUMMARY OF THE INVENTION

The present inventors earnestly investigated in order to develop a novel insecticide and consequently found that a hydrazinecarboxamide derivative represented by the general formula (I) shown below has an excellent insecticidal effect at a low dosage, whereby the present invention was accomplished.

That is, the present invention relates to a hydrazinecarboxamide derivative represented by the general formula (I):

wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkyl group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R⁴ is a hydrogen atom or a lower alkyl group, R³ and R⁴ being able to be taken together to represent an oxygen atom, A is

$$-N=C(-V)$$
)- or $-N(R^5)-CH(-V)$)-

(wherein X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower alkoxyalkyl groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, and R⁵ is a hydrogen atom; a lower alkylcarbonyl group; a lower alkoxydicarbonyl group; a cycloalkylcarbonyl group; a lower alkoxycarbonyl group; a lower alkoxydicarbonyl group; an unsubstituted phenylcarbonyl group; substituted phenylcarbonyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen

atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; or a substituted aminocarbonyl group having 1 or 2 substituents which may be the same or different and are selected from the group consisting of hydrogen atom, lower alkyl groups, unsubstituted phenyl group, and substituted phenyl groups having 1 to 5 substituents which may be the same or different and are selected from halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups), Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloakoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsulfonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylcarbonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom. The present invention further relates to a process for producing said derivative, and an agricultural and horticultural insecticide containing said derivative as an active ingredient.

In the definition of the substituents of the hydrazinecarboxamide derivative of the general formula (I), the term "lower" is used for expressing a number of carbon atoms of 1 to 6, and the prefix "halo" is used for expressing that a group has as its substituent(s) one or more halogen atoms which may be the same or different and are selected from the group consisting of chlorine atom, fluorine atom, bromine atom and iodine atom.

Of the hydrazinecarboxamide derivatives of the general formula (I) of the present invention, a hydrazinecarboxamide derivative of the genera formula (1') shown hereinafter has geometrical isomers, i.e., E-form and Z-form. The present invention also includes the E-form, the Z-form and mixtures comprising the E-form and Z-form in an arbitrary ratio. Each of hydrazinecarboxamide derivatives of the general formulas (I'') and (I''') shown hereinafter have stereoisomers, i.e., R-form and S-form. The present invention also includes the R-form, the S-form and mixtures comprising the R-form and the S-form in an arbitrary ratio.

All of the hydrazinecarboxamide derivatives of the general formulas (I'), (I") and (I'") are included in the hydrazinecarboxamide derivatives of the general formula (I).

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

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Preferable examples of the substituents of the hydrazinecarboxamide derivative of the above general formula (I) of the present invention are as follows. R¹, R², R³ and R⁴, which may be the same or different, are preferably hydrogen atoms; hydroxyl groups; or lower alkyl groups such as methyl, ethyl, n-propyl, i-propyl, i-butyl, i-butyl, etc. Preferable examples of substituent(s) for X are a hydrogen atom; halogen atoms such as chlorine, fluorine, bromine, iodine, etc.; a cyano group; lower alkyl groups such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, s-butyl, t-butyl, etc.; lower haloalkyl groups such as dichloromethyl, difluoromethyl, trichloromethyl, trifluoromethyl, difluoroethyl, tetrafluoroethyl, etc.; lower alkoxy groups such as methoxy, ethoxy, propoxy, etc.; and lower haloalkoxy groups such as

difluoromethoxy, trifluoromethoxy, difluoroethoxy, trifluoroethoxy, etc. As to the position(s) of the substituent(s) for X, at least one substituent is preferably at the 3-position.

Preferable examples of substituent(s) for Y are a hydrogen atom; halogen atoms such as chlorine, fluorine, bromine, iodine, etc.; a cyano group; a nitro group; alkyl groups such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, s-butyl, t-butyl, pentyl, heptyl, etc.; lower haloalkyl groups such as dichloromethyl, difluoromethyl, trifluoromethyl, etc.; lower alkylsulfonyloxy groups such as methyl sulfonyloxy, etc.; lower haloalkylsulfonyloxy groups such as trifluoromethyl sulfonyloxy, etc.; and lower alkoxycarbonyl groups such as methoxycarbonyl, etc. As to the position(s) of the substituent(s) for Y, at least one substituent is preferably at the 4-position. A particularly preferable example of the substituent(s) for Y is a cyano group.

Preferable examples of substituent(s) for Z are a hydrogen atom; halogen atoms such as chlorine, fluorine, bromine, iodine, etc.; a cyano group; alkyl groups such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, s-butyl, t-butyl, pentyl, heptyl, etc.; lower haloalkyl groups such as dichloromethyl, difluoromethyl, trichloromethyl, trifluoromethyl, trifluoroethyl, tetrafluoroethyl, etc.; lower alkoxy groups such as methoxy, ethoxy, propoxy, butoxy, etc.; lower haloalkoxy groups such as dichloromethoxy, difluoromethoxy, trichloromethoxy, trifluoromethoxy, difluoromethoxy, trifluoroethoxy, tetrafluoroethoxy, etc.; lower alkylthio groups such as methylthio, etc.; lower haloalkylthio groups such as difluoromethylthio, trifluoromethylthio, tetrafluoroethylthio, etc.; lower alkylsulfinyl groups such as methylsulfinyl, trifluoromethylsulfinyl, tetrafluoroethylsulfinyl, etc.; lower alkylsulfonyl groups such as methylsulfonyl, trifluoromethylsulfinyl, etc.; lower alkylsulfonyl groups such as methylsulfonyl, trifluoromethylsulfonyl, tr

R⁵ is preferably a hydrogen atom; a lower alkylcarbonyl group such as methylcarbonyl, ethylcarbonyl or the like; a lower haloalkylcarbonyl group such as difluoromethylcarbonyl, trifluoromethylcarbonyl, tetrafluoroethylcarbonyl, or the like; or a lower alkoxycarbonyl group such as methoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, or the like. W is preferably an oxygen atom. However, the substituents of the hydrazinecarboxamide derivative of the general formula (I) of the present invention are not limited to the above-exemplified atoms and groups.

As to typical processes for producing the hydrazinecarboxamide derivative of the general formula (I) of the present invention, said derivative can be produced, for example, by the production processes illustrated below.

Process A

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wherein R1, R2, R3, R4, X, Y, Z and W have the same meanings as those defined above.

A hydrazinecarboxamide derivative of the general formula (I') can be produced by reacting a compound of the general formula (VII) with a compound of the general formula (VI) in the presence of an inert solvent and in the presence or absence of a catalyst.

Reaction A-1.

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General formula (VIII) → general formula (I')

As the inert solvent used in this reaction, any inert solvent may be used so long as it does not markedly inhibit the progress of the reaction. There can be exemplified alcohols such as methanol, ethanol, propanol, butanol, etc.; halogenated hydrocarbons such as dichloromethane, chloroform, carbon tetrachloride, etc.; aromatic hydrocarbons such as benzene, toluene, xylene, etc.; nitriles such as acetonitrile, benzonitrile, etc.; ethers such as Methyl Cellosolve, diethyl ether, diglyme, dioxane, tetrahydrofuran, etc.; carboxylic acids such as acetic acid, etc.; dimethylacetamide; dimethyl sulfoxide; and water.

These inert solvents may be used singly or as a mixture thereof.

As the catalyst used in the reaction, there can be used, for example, inorganic acids such as hydrochloric acids, sulfuric acid and the like, or organic acids such as p-toluenesulfonic acid and the like.

As to the amount of the catalyst used, it is sufficient that the catalyst is present in the reaction system in an amount of 0.001 to 10% by weight based on the weight of the compound of the general formula (VIII).

Although the reactants are used in equimolar amount because the reaction is an equimolar reaction, either of them may be used in excess.

The reaction temperature may be properly chosen in the range of room temperature to the boiling range of the inert solvent used. The reaction is carried out preferably at 70° to 80°C.

Although the reaction time is varied depending on the degree of reaction, the reaction temperature, and the like, it may be chosen in the range of several minutes to 48 hours.

After completion of the reaction, the desired compound is isolated from a reaction solution containing the desired compound by a conventional method such as distilling-off of the solvent, solvent extraction, etc., and if necessary, purified by recrystallization, column chromatography, etc., whereby the desired compound can be produced.

Process B (when R1 is a hydrogen atom)

wherein R¹, R², R³, R⁴, X, Y, Z and W have the same meanings as those defined above, except that R¹ is not a lower alkyl group.

A hydrazinecarboxamide derivative of the general formula (I') can be produced by reacting a compound of the general formula (VIII) with a hydrazine derivative of the general formula (VIII) in the presence of an inert solvent and in the presence or absence of a catalyst to obtain a compound of the general formula (V), and reacting the compound (V) with a compound of the general formula (IV) in the presence of an inert solvent and in the presence or absence of a catalyst after or without isolating the compound (V).

Reaction B-1.

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General formula (VIII) → general formula (V)

As the inert solvent used in this reaction, there can be used the inert solvents exemplified for reaction A-1.

The kind and amount of the catalyst used in this reaction may be the same as in reaction A-1.

The hydrazine derivative of the general formula (VII) used in this reaction may be used in the form of either any of various salts or an aqueous solution having a suitable concentration.

As to the amount of the hydrazine derivative of the general formula (VII) used, the hydrazine derivative can be used in an amount equimolar with or larger than the amount of the compound of the general formula (VIII). Preferably, the amount is properly chosen in the range of 2 to 10 moles per mole of the compound of the general formula (VIII).

The reaction temperature may be properly chosen in the range of room temperature to the boiling range of the inert solvent used. The reaction is carried out preferably at 70° to 80° C.

Although the reaction time is varied depending on the degree of reaction, the reaction temperature and the like, it may be chosen in the range of several minutes to 48 hours.

After completion of the reaction, the desired compound is isolated from a reaction solution containing the desired compound by a conventional method such as distilling-off of the solvent, solvent extraction, etc., and if necessary, purified by recrystallization, column chromatography, etc., whereby the desired compound can be produced.

The compound of the general formula (V) obtained by the reaction may be subjected to the subsequent reaction either after isolation and purification by the above method, or without isolation.

Typical examples of the compound of the general formula (V) obtained by the present production process are listed in Table 1.

15				Physical properties	mp. 55°C	paste	mp. 91°C	mp. 72°C	paste	paste	paste
25		(v)		X	ш	4-F	4-CN	4-NO ₂	4 – F	Ħ	Ξ
30		E ^R − 2 − R − A − A − A − A − A − A − A − A − A	le 1	×	Ħ	×	Ħ	Ħ	4 된 -	Ħ	Ħ
35		$HN(R^2)-N=C$	Table	R4	н	#	#	Ħ	Ħ	#	æ
40) NH		ж ₃	Н	Ħ	Ħ	Ħ	Ħ	НО	#
45	cmula (V)			R ²	Ħ	Ħ	Ħ	н	н	н	CH ₃
50	neral formula			No.	V-1	V-2	V-3	V-4	V-5	N-6	V-7

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5	paste	paste	paste	paste	paste	paste	paste	paste	paste	paste	paste	
10	4-CN	4-C1	Ħ	3-C1	4-CH ₃	Ħ	Ħ	4-CN	4-CN	4-CN	4-CN	
cont'd)	3-C1	Ħ	4-N(CH ₃) ₂	3-C1	4-CH ₃	ж	ж	3-C1	3 - F	3-Br	3-CF3	
Table 1	# 	=	Ħ	ш	н	H	н	H	Ħ	н	H	
30	=	но	НО	НО	НО	CH ₃	OCH3	Ħ	#	Ħ	Ħ	
35 .	CH ₃	ĸ	Н	H	н	H	Ħ	н	н	н	Ħ	
40	V-8	0-V	V-10	V-11	V-12	V-13	V-14	V-15	V-16	V-17	V-18	

Table 2 shows 1-NMR data of the compounds having physical properties as paste listed in Table 1.

Table 2

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	No.	¹ H-NMR(CDCl ₃ /TMS, δ value, ppm.)
10	V-2	3.74+3.95(s, 2H), 5.13+5.33(bs, 2H),
		6.37-7.76(m, 9H).
15		(Mixture of E- and Z-forms)
	V-5	3.72+3.98(s, 2H), 5.10+5.37(bs, 2H),
		6.85-7.69(m, 8H).
20		(Mixture of E- and Z-forms)
	V-6	4.73(bs, 1H), 5.18(bs, 2H), 5.31(s, 1H),
25		6.71-7.58(m, 10H).
	V-7	2.98(s, 3H), 3.95(s, 2H), 5.00(bs, 1H),
30		6.90-7.81(m, 10H).
50	V-8	3.06(s, 3H), 4.00(s, 2H), 5.08(bs, 1H),
		7.22-7.69(m, 8H).
35	V-9	4.91(bs, 1H), 5.29(bs, 2H), 5.37(bs, 1H),
		6.85-7.45(m, 9H).
10	V-10	2.96(s, 6H), 4.96(bs, 1H), 5.31(bs, 2H),
J		5.39(bs, lH), 6.60-7.25(m, 9H).
	V-11	4.73(bs, lH), 5.32(bs, 2H), 6.18(bs, lH),
45		6.81-7.12(m, 8H).

- cont'd -

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Table 2 (cont'd)

5	V-12	2.27(s, 3H), 2.31(s, 3H), 4.81(bs, 1H),
		5.25(bs, 2H), 5.86(bs, 1H), 6.86-7.52(m, 8H).
10	V-13	1.50(d, 3H), 3.95(q, 1H), 4.98(bs, 2H),
		6.77-7.52(m, 10H).
15	V-15	3.82+4.06(s, 2H), 5.23+5.51(bs, 2H),
15		6.96-7.85(m, 8H).
		(Mixture of E- and Z-forms)
20	V-16	3.81+4.03(s, 2H), 5.26+5.55(bs, 2H),
		6.81-7.62(m, 8H),
25		(Mixture of E- and Z-forms)
	V-17	3.81+4.04(s, 2H), 5.22+5.51(bs, 2H),
		7.00-7.85(m, 8H).
30		(Mixture of E- and Z-forms)
	V-18	3.84+4.10(s, 2H), 5.21+5.58(bs, 2H),
35		7.24+7.96(m, 8H).
		(Mixture of E- and Z-forms)

Reaction B-2.

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General formula (V) → general formula (I')

As the inert solvent used in this reaction, there can be used, for example, the inert solvents usable in reaction A-1 except for the alcohols, the carboxylic acids and water. There can also be used esters such as ethyl acetate and the like and pyridines.

As the catalyst used in this reaction, there can be used, for example, amines such as triethylamine and the like. The amount of the catalyst used may be properly chosen in the range of catalytic amount to a number of moles larger than that of the compound of the general formula (V).

Although the reactants are used in equimolar amount because the reaction is an equimolar reaction, either of them may be used in excess.

The reaction temperature may be properly chosen in the range of -20°C to the boiling range of the inert solvent used, and is preferably in the range of -10°C to room temperature.

Although the reaction time is varied depending on the degree of reaction, the reaction temperature and the like, it may be chosen in the range of several minutes to 48 hours.

After completion of the reaction, the same treatment as in the case of reaction B-1 is carried out, whereby the desired compound can be produced.

Process C

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wherein R1, R2, R3, R4, X, Y, Z and W have the same meanings as those defined above.

A hydrazinecarboxamide derivative of the general formula (I") can be produced by reducing a hydrazinecarboxamide derivative of the general formula (I') with a reducing agent or by catalytic hydrogenation in the presence or absence of an inert solvent.

Reaction C-1.

General formula (I') → general formula (I")

This reduction reaction can be carried out by the use of a suitable reducing agent, or it can be carried 40 out as catalytic hydrogenation in the presence of a suitable catalyst.

As the reducing agent, there can be used, for example, metal hydrides such as LiAlH4, NaBH3CN, LiBH₃CN, etc. and reducing agents such as NaHSO₃ and the like. The amount of the reducing agent used may be chosen so that its number of moles in terms of the number of moles of hydride as reducing agent may be equal to or larger than that of the hydrazinecarboxamide derivative of the general formula (I').

As the inert solvent used in the reaction, any inert solvent may be used so long as it does not markedly inhibit the progress of the reaction. There can be exemplified alcohols such as methanol, ethanol, propanol, butanol, etc.; ethers such as diethyl ether, diglyme, dioxane, tetrahydrofuran, etc., Cellosolves such as Methyl Cellosolve, etc.; dimethylformamide; dimethylacetamide; dimethyl sulfoxide; sulfolane; and water.

These inert solvents may be used singly or as a mixture thereof.

The reaction is carried out under acidic or neutral conditions in the pH range of 1 to 7. The pH is preferably in the range of 4 to 6 and is adjusted by adding hydrogen chloride, hydrogen bromide or the like to the reaction system.

The reaction temperature may be properly chosen in the range of -20°C to the boiling range of the inert solvent used.

Although the reaction time is varied depending of the scale of reaction, the reaction temperature and the like, it is several minutes to 48 hours.

After completion of the reaction, the desired compound is isolated from a reaction solution containing

the desired product by a conventional method such as distilling-off of the solvent, solvent extraction, etc., and if necessary, purified by recrystallization, column chromatography, etc., whereby the desired compound can be produced.

When catalytic hydrogenation is carried out as the reduction reaction, it is carried out according to, for example, the conventional method described in Shin Jikken Kagaku Koza, Vol. 15-11, Maruzen Co., Ltd., etc. As the solvent usable in this case, there can be exemplified alcohols such as methanol, ethanol, propanol, butanol, etc.; Cellosolves such as Methyl Cellosolve, etc.; ethers such as diethyl ether, diglyme, dioxane, tetrahydrofuran, etc.; hydrocarbons such as hexane, cyclohexane, etc.; fatty acids or esters thereof, such as acetic acid, ethyl acetate, etc.; amides such as dimethylformamide, dimethylacetamide, N-methylpyrrolidone, etc., and ureas such as dimethylimidazoline, tetramethylurea, etc.

These inert solvents may be used singly or as a mixture thereof.

As the catalyst used in the reaction, there can be used typical catalysts for catalytic hydrogenation, for example, palladium-carbon, palladium black, platinum dioxide and Raney nickel. The amount of the catalyst used may be properly chosen in the range of 0.0001 to 20% by weight based on the weight of the hydrazinecarboxamide derivative of the general formula (I').

The hydrogen pressure in the reaction can be chosen in the range of atmospheric pressure to 300 atmospheres and is preferably in the range of atmospheric pressure to 50 atmospheres.

The reaction temperature may be properly chosen in the range of room temperature to the boiling range of the inert solvent used and is preferably in the range of room temperature to 80°C.

Although the reaction time is varied depending on the scale of reaction, the reaction temeprature and the like, it is several minutes to 80 hours.

After completion of the reaction, a reaction solution containing the desired compound is treated in the same manner as in the case of using the reducing agent, whereby the desired compound can be produced.

Process D

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wherein R¹, R², R³, R⁴, R⁵, X, Y and Z have the same meanings as those defined above, R^{5'} is a lower alkylcarbonyl group, a lower haloalkylcarbonyl group, a cycloalkylcarbonyl group, a lower alkoxycarbonyl group, an unsubstituted phenylcarbonyl group, or a substituted phenylcarbonyl group, or a substituted phenylcarbonyl group, an unsubstituted phenylcarbonyl group, or a substituted phenylcarbonyl group, an unsubstituted phenylcarbonyl group, and an unsubstituted phenylcarbonyl group, and an unsubstituted pheny

bonyl group having 1 to 5 substituents which may be the same or different and are selected from the group consisting of a halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, R^{5"} is a lower alkyl group, an unsubstituted phenyl group, or a substituted phenyl group having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups and Hal is a halogen atom.

A hydrazinecarboxamide derivative of the general formula (I''') can be produced by reacting a hydrazinecarboxamide derivative of the general formula (II') with a halide of the general formula (III) or an isocyanate of the general formula (III) in the presence of an inert solvent and a base.

Reaction D-1.

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General formula (I") → general formula (I'")

As the inert solvent used in this reaction

As the inert solvent used in this reaction, any inert solvent may be used so long as it does not markedly inhibit the progress of the reaction. There can be exemplified halogenated hydrocarbons such as methylene chloride, chloroform, carbon tetrachloride, etc.; aromatic hydrocarbons such as benzene, toleuen, xylene, etc.; esters such as ethyl acetate, etc.; nitriles such as acetonitrile, benzonitrile, etc.; ethers such as Methyl Cellosolve, diethyl ether, diglyme, dioxane, tetrahydrofuran, etc.; sulfolane; and dimethylsulfoxide. The inert solvents may be used singly or as a mixture thereof.

· As the base used in the reaction, inorganic based or organic bases can be used. As the inorganic bases, there can be exemplified hydroxides, carbonates and alcoholates of alkali metals or alkaline earth metals, for example, sodium, potassium, magnesium and calcium. As the organic bases, triethylamine and pyridine can be exemplified.

Although the reactants are used in equimolar amount because the reaction is an equimolar reaction, either of them may be used in excess.

The reaction temperature may be chosen in the range of 0°C to the boiling range of the inert solvent used.

Although the reaction time is varied depending on the scale of reaction, the reaction temperature and the like, it may be chosen in the range of several minutes to 48 hours.

After completion of the reaction, the desired compound is isolated from a reaction solution containing the desired compound by a conventional method and if necessary, purified by recrystallization, column chromatography, etc., whereby the desired compound can be produced.

Typical examples of the hydrazinecarboxamide derivative of the general formula (I) of the present invention are given below but they are not intended in any way to limit the scope of the present invention.

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20	5 − (1) 		
25	$) \longrightarrow N(R^{1}) - C - N(R^{2}) - N = C - C$	(1.)	Table 3
30)-N(R ¹)-	•	Ta
35	2		
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No.	R	R ²	п3	4	×	X	Z	W	Physical properties	l ies
A001	Н	Н	Н	н	Н	Н	н	0	m.p. 181°C	1°C
A002	н	Ħ	I	Ħ	Ħ	ж	2-c1	0	m.p. 19	197°C
A003	н	н	ж	Ħ	Ħ	ш	3-c1	0	m.p. 18	188°C
A004	H	н	н	н	Ħ	н	4-C1	0	m.p. 19	199°C
A005	Œ	H	Ħ	Ħ	Ħ	ж	4-0CH ₃	0	m.p. 14	144°C
A006	Ħ	H	н	н	H	н	4-OCHF ₂	0	m.p. 17	172°C
A007	н	H	н	Ħ	Ħ	H	4-0CF ₃	0	m.p. 14	149°C
A008	н	Ħ	н	Ħ	Ħ	4-0H	4-C1	0	m.p. 206°C	၁့9

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5		m.p. 211°C	m.p. 192°C	m.p. 224°C	m.p. 227°C	m.p. 204°C	m.p. 203°C	m.p. 209°C	m.p. 191°C	m.p. 206°C	m.p. 193°C	m.p. 186°C	m.p. 197°C	m.p. 217°C	m.p. 209°C	
10		0	0	0	0	0	0	0	0	0	0	0	0	0	0	_
15			8	-		က			ຸຕ			H ₇ -t	ຸຕ	$4-0$ CF $_2$ CHF $_2$	4-0-(O)-CF ₃	
20	, d)	4-CF3	4-0CF ₃	4-C1	4-CF3	4-0CF ₃	4-C1	4-CF3	4-0CF3	4-C1	4-CF3	4-0C4H7-t	4-0CF3	4-0CF	4-0-4	
25	(cont'd)															
30	Table 3	4-0H	4-0н	2-C1	2-C1	2-C1	3-C1	3-C1	3-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1	
35	H	Н	Ħ	Ħ	Ħ	×	H	×	Ħ	正	缸	н	ж	н	ш	
40		н	Ħ	Ħ	н	Ħ	н	н	ж	Ħ	ж	#	#	H	Ħ	
40		н	# 	#	#	#	H	н	Ħ	н	#	#	#	Ħ		
45		Н	Ħ	н	Ħ	н	ĸ	Ħ	Ħ	Ħ	Ħ	H	н	Ħ	Ħ	
		王	Œ	H	Ħ	ı	Ħ	Ħ	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ	
50		A009	A010	A011	A012	A013	A014	A015	A016	A017	A018	A019	A020	A021	A022	

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5		. 202°C	. 219°C	. 201°C	. 222°C	. 202°C	. 208°C	. 213°C	. 205°C	. 187°C	. 192°C	. 181°C	. 195°C	. 209°C	. 116°C	. 180°C
		m.p.	m.p.	·ď·m	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.
10		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15																
20	ļ	4-C1	4-CF3	4-0CF ₃	4-C1	4-CF3	4-0CF3	4-C1	4-CF3	4-0CF3	4-C1	4-CF ₃	4-0CF ₃		2-C1	3-C1
	(cont'd)	4-	4	4-	4-	4-	4-	4-	4-	4-	4-	4-	4-	H	2-	3-
25	(con															
	Table 3	2,4-Cl ₂	2,4-Cl ₂	2,4-Cl ₂	4-Br	4-Br	Br	ĮŢ.	۲	[II4	3-CN	3-CN	3-CN	4-CN	4 CN	4 –CN
30	Tabl	5	2,	2,	4	4	4-Br	4-F	요 1	4-F	e e	ا	<u>ه</u>	4-	4-	4-
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		I	耳	二	耳	#	H	#		H	H	H				
50		A023	A024	A025	A026	A027	A028	A029	A030	A031	A032	A033	A034	A035	A036	A037

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5		m.p. 217°C	m.p. 128°C	m.p. 230°C	m.p. 205°C	m.p. 208°C	m.p. 200°C	m.p. 207°C	m.p. 195°C	m.p. 231°C	m.p. 215°C	m.p. 213°C	m.p. 193°C	m.p. 217°C	m.p. 116°C	m.p. 204°C
10		0	လ	0	0	0	0	0	တ	0	0	0	0	0	Ø	0
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20	۲ . ۵)	4-C1	4-C1	3,4-C1 ₂	3,5-C1 ₂	4-Br	3-F	4-F	4-CN	4-NO2	4-CH ₃	4-C4H7-t	3-CF3	4-CF3	4-0CF3	4-0CH ₃
25	Table 3 (cont'd)															
30	rable 3	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN
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		#	#	H	H	H	H	H	H	=	Ξ	#	H	<u> </u>	<u> </u>	=
40		н	Ħ	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ	н
45		Ħ	#	Ħ	H	н	н	н	æ	×	æ	æ	Ħ	Ħ	Ħ	H
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50		A038	A039	A040	A041	A042	A043	A044	A045	A046	A047	A048	A049	A050	A051	A052

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5		m.p. 204°C	m.p. 197°C	Paste	m.p. 214°C E-form	m.p. 159°C Z-form	m.p. 221°C	m.p. 193°C	m.p. 206°C	m.p. 182°C	m.p. 215°C
10		0	0	ß	0	0	0	0	0	0.	0
15		tt.					3	F2		⊢ CF₃	⊢ CF ₃
20	d)	4-0C ₄ H ₇ -t	4-OCHF2	4-0CHF ₂	4-0CF ₃	4-0CF ₃	$4-0$ CH $_2$ CF $_3$	$4-0$ CF $_2$ CHF $_2$	4-0-	4-0-	4-0-
25	(cont'					-					
30	Table 3 (cont'd)	4-CN	4-CN	4-CN	4 - CN	4-CN	4-CN	4 -CN	4-CN	4-CN	4-CN
35	ç.	Ħ	н	Н	æ	Ħ	H	Ħ	н	Ħ	н
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50		A053	A054	A055	A056	A057	A058	A059	A060	A061	A062

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5		p. 167°C	. 216°C	. 228°C	p. 140°C	p. 230°C	p. 203°C	p. 195°C	p. 194°C	p. 220°C	p. 182°C
		m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.
10		0	0	0	0	0	0	0	0	0	0
15		.cr ₃	CF ₃	o-O-CF3							
20		O _z		3,5-Cl ₂ -4-0-	C1 3,5-C1 ₂ -4-0-	4-C00CH ₃	بر ج	4-0CF3		æ	_
	<u>-</u>	4-0-4	C1.	3,5-	3,5-	4-C	4-CF3	4-0(H	2-C1	3-C1
25	(cont'd)							-			
30	Table 3	4 -CN	4-CN	4-CN	4 -CN	4 CN	$2-NO_2$	$2-NO_2$	4-NO ₂	4-NO ₂	4-NO ₂
35	Tē	ж	*	ш	ж	#	H	H	H	×	æ
		-									
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50		A063	A064	A065	A066	A067	A068	A069	A070	A071	A072

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5		222°C	206°C	206°C	197°C	205°C	209°C	189°C	139°C	191°C	200°C	204°C	192°C	205°C	204°C	186°C	
5		m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p. 186°C	
10		0	ß	0	0	0	0	0	ຜ	0	0	0	0	0	0	0	
70																	
15										$^{ m IF}_2$							
20					ຸຕ	ູຕ	HF2	F.3	F 3	4-OCF2CHF2	Э			ູຕ	ູຕ	म् ह	
20	(p.	4-C1	4-C1	4-F	3-CF3	4-CF3	4-0CHF ₂	4-0CF3	4-0CF3	4-00	4-SCF ₃	4-C1	4-F	3-CF3	4-CF3	4-0CF3	
25	(cont'd)																
30	Table 3	4-NO ₂	4-NO ₂	$4-NO_2$	$4-NO_2$	4-NO ₂	4-NO ₂	4-NO ₂	$4-NO_2$	4-NO ₂	4-NO ₂	4-CH ₃					
	T.	-															
35		≖	#	H	エ	Ħ	Œ	H	H	田	田	Ħ	#	Ħ	H	#	_
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50		A073	A074	A075	A076	A077	A078	A079	A080	A081	A082	A083	A084	A085	A086	A087	

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	H H 4-0CH ₃ 4-Cl 0 m.p. 191°C	н	H H 4-0CH ₃	H H 4-0CHF2	H H 4-0CF ₂ CHF ₂ 4-0CF ₃ 0 m.p. 161°C	н	H H 4-0CH ₂ CH=CH ₂	H H 4-0-COCH ₃	H H 4-0SO ₂ CH ₃	H H 4-0S0 ₂ CF ₃	H H 4-SCH ₃	H H 4-SCH ₃	H H 4-SOCH ₃	H H 4-SOCH ₃ 4-OCF ₃ 0 m.p. 192°C	H H H 4-SO ₂ CH ₃ 4-C1 0 m.p. 210°C	
£∓ 35		표	H								 		····		<u></u>	
4 5 50	А088 Н Н	А089 Н Н	А090 Н Н	А091 Н Н	А092 Н Н	А093 Н Н	A094 H H	А095 Н Н	А096 Н Н	A097 H H	А098 Н Н	А099 Н Н	Aloo H H	A101 H H	A102 H H	_

EP 0 462 456 A1

5		J.96T	148°C	206°C	210°C	171°C	177°C	166°C	226°C	212°C	207°C	201°C	207°C	ງ . 561	183°C	183°C
		m.p.	m.p.	m.p.	d m	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.
10		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15																5-F5
20	·d)	4-0CF3	4-0CF3	4-0CF3	4-0CF3	4-C1	4-CF3	4-0CF3	4-CF3	4-0CF3	4-CF3	4-0CF3	4-C1	4-CF3	4-0CF3	2,3,4,5,6-F ₅
25 30	Table 3 (cont'd)	4-so ₂ cH ₃	4-NH ₂	4-NHCOCH ₃	4-NHSO ₂ CH ₃						4-c1	4-C1	4-F	Ę	Fi.	4-CN
30	Tab	4	4-	4-	4	皿	#			=	4	4	4	4 - A	4-F	4
35		Œ	н	·	H	2-C1	2-c1	2-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1
40		Ħ	#	Ħ	#	#	#	=	Ħ	Ħ	Ħ	Ħ	Ħ	H	Ħ	I
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50		A103	A104	A105	A106	A107	A108	A109	A110	A111	A112	A113	A114	A115	A116	A117

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	206°C	218°C	218°C	221°C	186°C	233°C	215°C	197°C	226°C	207°C	189°C	198°C	167°C	m.p. 187°C	E-form
	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	Ξ-
	0	0	0	0	0	0	0	0	0	0	0	0	ß	0	
(cont'd)	4-C1	2,3-C1 ₂	2,4-C1 ₂	2,5-C1 ₂	2,6-Cl ₂	3,4-C1 ₂	3,5-C1 ₂	4-Br	$4-C_4H_9-t$	4-CF3	4-0CH ₃	$4-0C_4H_9-t$	4-0CHF ₂	4-0CF3	
									·						
	4-CN	4-CN	4-CN	4-CN	4-CN	4 CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	
	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	
	Ħ	æ	Ħ	Ħ	Ħ	Ħ	Ħ	H	Ħ	H	н	н	н	н	
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	A118	A119	A120	A121	A122	A123	A124	A125	A126	A127	A128	A129	A130	A131	
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5		m.p. 148°C Z-form	m.p. 199°C	m.p. 226°C	m.p. 184°C	m.p. 207°C	m.p. 196°C	m.p. 184°C	m.p. 201°C	m.p. 198°C	m.p. 216°C	m.p. 196°C
10		0	Ω	0	0	0	0	0	0	0	0	0
15				$c_{{f F}_3}$	CHF ₂	$3,5$ - cl_2 -4- 0 CF $_2$ CHF $_2$	CHClF	CHFCF3	\bigcirc	\bigcirc -cr ₃	\rightarrow cF ₃	\sim CF ₃
20	'd)	4-oce3	4-0CF ₃	4-0CH ₂ CF ₃	4-OCF2CHF2	3,5-C1	4-OCF2CHCIF	4-OCF2CHFCF3	4-0-	4-0-	15 0-4 O	4-0-{ N
25	(cont'd)											
30	Table 3	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN
35		3-C1	3-C1	3-C1	3-C1	3-C1	3-01	3-C1	3-c1	3-C1	3-c1	3-C1
		Ħ	Ħ	н	н	Ħ	Ħ	н	æ	×	×	H .
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45		Ħ	н	Ħ	н	H	н	H	Ħ	Ħ	Ħ	Ħ
		#	н	=	¤.	Ħ	Ħ	Ħ	異	Ħ	エ	E
50		A132	A133	A134	A135	A136	A137	A138	A139	A140	A141	A142
50	ι											

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5		m.p. 223°C	m.p. 206°C	m.p. 145°C	m.p. 194°C	m.p. 215°C	m.p. 195°C	m.p. 221°C	m.p. 216°C	m.p. 205°C	m.p. 217°C	m.p. 253°C
		0	0	0	0	0	0	0	0	0	0	0
10)- CF3) - cr3								
15)-cr3	2-4-0-	C1 C1			8	CHF2	m	m	$^{\mathrm{CHF}_2}$.e.
20	,q)	4-0-4	3,5-C1 ₂ -4-0	Cl. 3,5-Cl ₂ -4-0→	4-SCH ₃	4-SCF ₃	4-SCHF2	4-SCF ₂ CHF ₂	4-SOCH ₃	4-SOCF3	4-SOCF ₂ CHF ₂	4-SO ₂ CH ₃
25	Table 3 (cont'd)											
30	rable	4-CN	4-CN	4-CN	4-CN	4 -CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN
35		3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1
		Ħ	I	H	н	Ħ	Ħ	Ħ	Ħ	=	×	н
40		##	=	Ħ	н	Ħ	Ħ	=	#	н	H	ж
45		ж	Ħ	н	н	Ħ	н	Ħ	Ħ	Ħ	Ħ	H
		Ħ	Ħ	Ħ	Ξ	Ħ	H	Ξ:	Ħ	Ħ	H	Ħ
50		A143	A144	A145	A146	A147	A148	A149	A150	A151	A152	A153

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5	m.p. 212°C	m.p. 212°C	m.p. 212°C	m.p. 216°C	m.p. 211°C	m.p. 192°C	m.p. 210°C	m.p. 184°C	m.p. 190°C	m.p. 201°C	m.p. 195°C	m.p. 174°C	m.p. 221°C	m.p. 216°C	m.p. 182°C
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15															
20	4-SO ₂ CHF ₂	$4-SO_2CF_3$	4-SO2CF2CHF2	4-C00CH ₃	4-CF3	4-0CF ₃	11	4-CF ₃	4-0CF ₃	:1	4-CF ₃	4-0CF ₃	:1	4-CF ₃	4-0CF ₃
'd)	4-8	4-6	4-5	4-(4-(4-0	4-C1	4-0	4-0	4-C1	4-0	4-0	4-C1	4-0	4-c
Gont'd)															
S Table 3	4-CN	4-CN	4-CN	4-CN	H	Ħ	4-C1	4-C1	4-C1	4-F	4-F	4-F	4-CN	4-CN	4-CN
35	3-C1	3-C1	3-C1	3-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1	4-C1
	=	н	×	=	=	æ		н	Ħ	—	*	=	H	Ħ	H
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	#	<u> </u>	H	H	=	=	=	==	H	=	H	=	=	=	E
50 ·	A154	A155	A156	A157	A158	A159	A160	A161	A162	A163	A164	A165	A166	A167	A168

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40	40			00	35	25	15	10	5	
				i	Ŧ	Table 3 (cont'd)	q)			
н	н		н		4-C1	4-CN	4-0CF3	ß	m.p. 193°C	
н н н	æ				4-C1	4-CN	$4-0$ CF $_2$ CHF $_2$	0	m.p. 201°C	
н н н	н	н			4-C1	4-NO ₂	4-C1	0	m.p. 217°C	
н	H	Ħ		4.	4-C1	4-NO ₂	4-CF ₃	0	m.p. 213°C	
н н н н	н	н		ব	4-C1	4-NO ₂	4-0CF ₃	0	m.p. 206°C	
н н н н з	н	Ħ		က	3,4-C1 ₂	н	4-C1	0	m.p. 212°C	
н н н н з	н	Ħ		സ	3,4-Cl ₂	H	4-CF ₃	0	m.p. 206°C	
н н н н з	н	ш		က	3,4-C1 ₂	н	4-0CF ₃	0	m.p. 193°C	
н н н н	н	Ħ		ന	3,4-C1 ₂	4-CN	4-C1	0	m.p. 226°C	
н	н	Ħ		(.)	3,4-C1 ₂	4-CN	4-CF ₃	0	m.p. 212°C	
н	н	×		.,	3,4-C1 ₂	4-CN	4-0CF ₃	0	m.p. 193°C	
ннн	н	耳		(r)	3,5-C1 ₂	H	4-C1	0	m.p. 239°C	
н н н з	н	Ħ		സ	3,5-C1 ₂	н	4-CF ₃	0	m.p. 227°C	
ннн	H	E		(.)	3,5-C1 ₂	н	4-0CF ₃	0	m.p. 227°C	
н	н	H		(.)	3,5-C1 ₂	4-CN	4-C1	0	m.p. 228°C	
			_	_ j				_		

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5	213°C	206°C	191°C	228°C	209°C	205°C	223°C	233°C	194°C	220°C	197°C	176°C	183°C	186°C	188°C
	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
٠												_			
15							7	7	7	7			CHF ₂	CHCLF	CHFCF ₃
20 (p	4-CF3	4-0CF3	4-C1	4-CF ₃	4-0CF ₃	4-C1	2,3-C1 ₂	2,4-C1 ₂	2,6-C1 ₂	3,4-C1 ₂	4-CF3	4-0CF3	4-0CF2CHF2	4-OCF2CHC1F	4-OCF2CHFCF3
25 (cont'd)											•				
s Table 3	4-CN	4-CN	H	н	Ħ	4-CN	4 -CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN
35	3,5-C1 ₂	3,5-C1 ₂	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	3-Br
	Ħ	H	#	Ħ	Ħ	Ħ	Ħ	×	Ħ	Ħ	æ	H	Ħ	Ħ	н
40	ж	н	Ħ	H	Ħ	н	Ħ	H	Ħ	н	H	н	#	Ħ	I
45	Ħ	н	H	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ	н	Ħ	н	æ	ж
	Ħ	≖	Ħ	エ	Ħ	耳	Ξ	H	五	H	Ħ	Ħ	H	H	H
50	A184	A185	A186	A187	A188	A189	A190	A191	A192	A193	A194	A195	A196	A197	A198

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5		m.p. 218°C	m.p. 206°C	m.p. 221°C	m.p. 216°C	m.p. 193°C	m.p. 215°C	m.p. 204°C	m.p. 192°C	m.p. 197°C	m.p. 175°C	m.p. 177°C	m.p. 173°C	m.p. 180°C	m.p. 196°C	m.p. 170°C
10	-	0	0	0	0	0	0	0	0	0	0	0	0	0	<u> </u>	0
15		8		2	æ	$^{ m HF}_2$	я 3									
20	d)	4-SCHF ₂	4-SCF3	4-SOCHF ₂	4-SOCF ₃	$4-SO_2CHF_2$	$4-SO_2CF_3$	4-C1	4-CF3	4-0CF3	4-C1	4-CF ₃	4-0CF ₃	4-C1	4-CF ₃	4-0CF ₃
25	Table 3 (cont'd)	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	3,4-(OCH ₃) ₂	3,4-(OCH ₃) ₂	3,4-(OCH ₃) ₂				4-CN	4-CN	4-CN
30	Tat	4	4	4	4	4	4	<u>. </u>	<u>~</u>	<u>~</u>	=	=	#	4		4
35		3-Br	3-Br	3-Br	3-Br	3-Br	3-Br	4-Br	4-Br	4-Br	2-F	2-F	2-F	2-F	2~F	2-F
		Ħ	Ħ	H	H	н	Ħ	H	H	#	Ħ	×	Ħ	Ħ	Ħ	Ħ
40		Ħ	×	н	н	Ħ	Ħ	Ħ	н	н	н	н	Ħ	H	Ħ	н
4 5		×	×	Ħ	H	н	Ħ	H	н	Ħ	Ħ	H	=	Ħ	Ħ	н
40	ļ	=======================================	H	Ħ	Ħ	×	Ħ	Ħ	Ħ	Ħ	H	Ħ	×	×	エ	H
50		A199	A200	A201	A202	A203	A204	A205	A206	A207	A208	A209	A210	A211	A212	A213

5		206°C		200°C	209°C	.191°C	208°C	216°C	202°C	203°C	191°C	222°C	185°C	184°C	199°C	178°C
		m. p.		m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	n.p.
10		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15			•													
20	,q)	4-C1	4-CF3	4-0CF3	4-CF3	4-0CF3	4-C1	4-CF3	4-0CF3	4-C1	4-0CF3	4-C1	4-CF ₃	4-0CF ₃	4-CF3	4-0CF3
25	3 (cont'd)															
30	Table 3	Ħ	н	н	4-C1	4-C1	4-CN	4 -CN	4 -CN	Ħ	Ħ	4 -CN	4-CN	4-CN	4 - F	4 - F
35	•	3-F	3-F	3-F	3-F	3-F	3-F	3-F	3-F	4-F	4-F	4-F	4-F	4-F	4-F	4-F
		Н	н	н	н	Ħ	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	æ
40		н	Ħ	H	Ħ	#	Ħ	н	Ħ	н	×	×	×	ж_	Ħ	×
45		н	H	H	H	Ħ	ж	H	Ħ	H	H	≖	×	æ	#	=
		н	Н	Ħ	н	Ħ	Ħ	Ħ	н	н	н	Ħ	H	Ħ	Ħ	Ħ
50		A214	A215	A216	A217	A218	A219	A220	A221	A222	A223	A224	A225	A226	A227	A228
														_		

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50		45	40		35	25	15	10		5	
					H	Table 3 (cont'd)	ď)				
A229	Н	Н	Н	Н	4-F	4-F	4-0CF ₂ CHF ₂	0	m.p.	187°C	
A230	Ħ	æ	Ħ	Ħ	4-F	4-CN	4-C1	0	m.p.	232°C	
A231	Ħ	Ħ	#	Ħ	4-F	4-CN	4-CF3	0	m.p.	202°C	
A232	二.	H	н	H	4-F	4-CN	4-0CF ₃	0	m.p.	210°C	
A233	Ħ	æ	Ħ	×	4-F	4-NO ₂	4-C1	0	m.p.	209°C	
A234	Ξ	Ħ	H	H	4-F	4-NO ₂	4-CF3	0	m.p.	220°C	
A235	H	н	Ħ	Ħ	4-F	4-NO ₂	4-ocr3	0	m.p.	204°C	
A236	H	H	Ħ	H	3,5-F2	н	4-C1	0	m.p.	218°C	
A237	Ħ	ж	н	I	3,5-F2	H	4-CF3	0	m.p.	208°C	
A238	Ħ	н	Ħ	Ħ	3,5-F2	н	4-ocF ₃	0	m.p.	211°C	
A239	缸	Ħ	Ħ	Ħ	3-I	4-CN	4-C1	0	m.p.	213°C	
A240	Ħ	Ħ	Ħ	Ħ	3-I	4-CN	4-CF ₃	0	m.p.	205°C	
A241	Ħ	н	н	Ħ	3-I	4-CN	4-0CF3	0	m.p.	201°C	
A242	Ħ	Ħ	Ħ	H	3-I	4-CN	$4-0$ CF $_2$ CHF $_2$	0	m.p.	2°961	
A243	Ħ	Ħ	Ħ	×	2-сн ₃	н	4-C1	0	m.p.	121°C	

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8		_															
H H H H H 3-CF3	5		m.p. 210°C	m.p. 188°C	m.p. 191°C	m.p. 149°C											m.p. 238°C
H H H 3-CF ₃ H 4-OCF ₃ H H H 3-CF ₃ 4-CN 4-CCF ₃ H H H 3-CF ₃ 4-CN 4-CCF ₃ H H H 3-CN 4-CN 4-CCF ₃ H H H 3-CN 4-CN 4-CCF ₃ H H H 3-CN 4-CN 4-CCF ₃ H H H 4-CN 4-CN 4-CCF ₃ H H H 4-CN 4-CN 4-CCF ₃ H H H 4-CN 4-CN 4-CCF ₃ H H H H 4-CN 4-CN 4-CCF ₃ H H H H 4-CN 4-CN 4-CCF ₃ H H H H 4-CN 4-CN 4-CCF ₃	10	}	0				0	0									
H H H H 3-CF ₃	15		3		m	m	$_2^{CHF}_2$	m	m					e			3
H H H H 3-CF ₃	20		4-0CF	4-CF3	4-0CF	4-0CF	4-0CF	4-0CF	4-0CF	4-C1	4-CF3	4-C1	4-CF3	4-0CF	4-C1	4-CF3	4-0CF
Tabl H H H H H 3-CF3 H H H H H 3-CF3 H H H H H 3-CF3 H H H H 3-CF3 H H H H 4-CN H H H H 4-CN H H H H H 4-CN H H H H 4-CN										-							
40			3-CF3	3-CF3	3-CF3	3-CF3				3,5-(CF ₃) ₂	3,5-(CF ₃) ₂	3-CN	3-CN	3-CN	4-CN	4-CN	4-CN
			Ħ	Ħ	H		H	Ħ	H	Ħ	Ħ	H	æ	Ħ	H		Ħ
	40	-	н	Ħ	н	E	Н	Ħ	н	н	Ħ	н	×	Ħ	=	Ħ	Н
	45		H	н	н	н	н	н	H	×	×	×	Ħ	Ħ	Ħ	Œ	Н
A259 A260 A261 A261 A263 A264 A265 A266 A266 A267 A269 A270 A270		Ī	I	×	Ħ	×	Ħ	Ħ	Ħ	Ħ	Ħ	H	王	王	Ħ	耳	Ξ
	50		A259	A260	A261	A262	A263	A264	A265	A266	A267	A268	A269	A270	A271	A272	A273

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5		m.p. 194°C	m.p. 201°C	m.p. 217°C	m.p. 210°C	m.p. 177°C	m.p. 180°C	m.p. 182°C	m.p. 168°C	m.p. 171°C	m.p. 185°C	m.p. 182°C	m.p. 188°C	m.p. 194°C	m.p. 204°C	m.p. 202°C
10		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15		.F.3		ຼຸຕ	Т.3	_ m	Н.3		့်က	F3		.F.3	1F3		ຼຸຕ	F3
20		4-0CF3	4-C1	4-CF ₃	4-0CF ₃	4-CF3	4-0CF3	4-C1	4-CF3	4-0CF3	4-C1	4-0CF3	4-0CF ₃	4-C1	4-CF3	4-0CF ₃
25	Table 3 (cont'd)	4-CN	н	H	H	4-CN	4-CN	4-CN	4-CN	4-CN	×	н	4-CN	H	×	Ħ
30 35	Tab]	3-0CH ₃	4-0CH ₃	4-0CH ₃	4-0CH ₃	3-0C3H7-i	3-0C3H7-i	3-0-6	3-0-6	3-0-6	3-OCHF2	3-OCHF2	3-OCHF2	4-ochF ₂	4-OCHF ₂	4-OCHF ₂
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40		Ħ	H	×	×	ж	H	Ħ	×	Ħ	Ħ	ĸ	Ħ	±	×	H
45		Ħ	щ	Ħ	Ħ	Ħ	Ħ	Ħ	I	н	Ħ	ж	Ħ	Ħ	. #	H
		田	Ħ	H	I	#	H	Ħ	H	Ξ	Ħ		Ξ	H	Ħ	#
50		A274	A275	A276	A277	A278	A279	A280	A281	A282	A283	A284	A285	A286	A287	A288

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5	. 213°C	. 208°C	. 204°C	. 175°C	. 206°C	. 182°C	. 180°C	. 211°C	. 200°C	. 199°C	. 195°C	. 113°C	. 132°C	. 108°C
	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.
10	0	0	0	0	0	0	0		0	0	0	0	0	0
15	4-OCF ₂ CHF ₂	4-SCF ₃	4-SOCF ₃	4-OCF ₂ CHF ₂	4-C1	4-CF ₃	4-0CF3	4-C1	4-CF ₃	4-0CF ₃	4-oce ₂ che ₂	н	4-C1	4-oce3
20														
(cont.d)	Ħ	Ħ	H	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	н	æ	н
s Table 3	4-OCHF ₂	$4-0$ CHF $_2$	$4-0$ CHF $_2$	$4-0$ CF $_2$ CHF $_2$	3-0CH ₂ 0-4	3-0CH ₂ 0-4	3-0CH ₂ 0-4	2-CH=CHCH=CH-3	2-CH=CHCH=CH-3	2-сн=снсн=сн-3	2-СН=СНСН=СН-3	н	н	Н
35	4.	<u>4</u> ,	4	<u> </u>		<u></u>	(1)	~				<u></u>		
	<u> </u>	Ξ_	H	H	#	H	#	Ħ	#	#	<u> </u>	Ξ_	耳	Ħ
40	Ħ	Ħ	Ħ	田	H	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	H
	#	Ħ	н	Ħ	Ħ	н	н	Ħ	Ħ	Ħ	#	Ħ	Ħ	н
45	×	H	H	Ħ	Ħ	H	H	H	Ħ	Ħ	Ħ	СНЗ	CH ₃	СНЗ
50	A289	A290	A291	A292	A293	A294	A295	A296	A297	A298	A299	A300	A301	A302

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5		m.p. 111°C	m.p. 117°C	m.p. 108°C	m.p. 98°C	Paste	m.p. 85°C	Paste	m.p. 148°C	Paste	m.p. 115°C EZ-form	m.p. 95°C E-form	m.p. 66°C Z-form	m.p. 121°C
10		0	0	0	0	0	0	0	0	0	0	0	0	0
15														
73		н	2-C1	3-C1	4-C1	$3,4-C1_2$	4-Br	4-CH ₃	4-CF3	4-0CH ₃	4-0CF3	4-0CF ₃	4-0CF3	4-C1
20														
25	Table 3 (cont'd)	出	×	×	ш	H	I	E	Ħ	Ħ	Ħ	Œ.	н	4-C1
30	Table 3													
35		H	Ħ	H	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	H	Ħ	H	Ħ
		エ	н	×	Ħ	Ħ	×	Ħ	æ	æ	Ħ	н	Ħ	н
40		H	Ħ	Ħ	H	Ħ	Ħ.	Ħ	=	Ħ	Ħ	Ħ	Ħ	н
		CH ₃	СН3	СН3	CH ₃	СНЗ	СНЗ	СН3	сн3	CH ₃	СНЗ	CH ₃	СНЗ	СНЗ
45		Ħ	Ħ	H	H	Ħ	Ħ	H	Ħ	Ħ	Ħ	×	н	н
		33					80	60	01	=======================================		13	14	15
50		A303	A304	A305	A306	A307	A308	A309	A310	A311	A312	A313	A314	A315

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	Paste	Paste	m.p. 105°C	m.p. 140°C	m.p. 127°C	m.p. 98°C	Paste	Paste	m.p. 109°C	m.p. 119°C	Paste	Paste	Paste	Paste	m.p. 167°C
	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
·	3,4-C1 ₂	3-CF3	4-0CF ₃	4-C1	4-CF3	4-0CF3	2-C1	3-C1	4-C1	4-CF3	4-0CF ₃	4-0CF ₃	4-C1	4-0CF3	н
	4-C1	4-C1	4-C1	4-CN	4-CN	4 -CN	Ħ	н	Ħ	#	H	Ħ	Ħ	Ħ	н
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	CH ₃	CH ₃	CH ₃	CH ₃	CH ₃	CH ₃	CH ₃	CH ₃	CH ₃	СН3	CH ₃	СН3	$_3$	CH ₃	Ħ
Ì	×	Œ	Ħ	エ	Ħ	Ħ	×	Ħ	Ħ	H	Ħ	Ħ	H	×	Ħ
	A316	A317	A318	A319	A320	A321	A322	A323	A324	A325	A326	A327	A328	A329	A330
		H CH ₃ H H H 4-C1 3,4-C1 ₂ 0	H CH ₃ H H H 4-C1 3,4-C1 ₂ 0 H CH ₃ H H H 4-C1 3-CF ₃ 0	H CH ₃ H H H 4-Cl 3,4-Cl ₂ 0 Paste H CH ₃ H H H 4-Cl 3-CF ₃ 0 Paste H CH ₃ H H H 4-Cl 4-Cl 0 m.p.	H CH ₃ H H H 4-Cl 3.4-Cl ₂ O Paste B 4-Cl 3-CF ₃ O Paste H CH ₃ H H 4-Cl 4-Cl 0 Paste H CH ₃ H H A-Cl 4-Cl 0 m.p.	H CH ₃ H CH H H H CH H H H CH H H CH H H H CH H H H CH H H H H CH H	H CH ₃ H H H H H H CH ₃ H H H H H H H H H CF ₃ O Paste H CH ₃ H H H H-CI 4-CI 0 m.p. H CH ₃ H H 3-CI 4-CN 4-CI 0 m.p. H CH ₃ H H 3-CI 4-CN 4-CK ₃ O m.p. H CH ₃ H H 3-CI 4-CN 4-CK ₃ O m.p.	H CH ₃ H H H 4-C1 3,4-C1 ₂ 0 Paste H CH ₃ H H H 4-C1 3-CF ₃ 0 Paste H CH ₃ H H H 4-C1 4-OCF ₃ 0 m.p. H CH ₃ H H 3-C1 4-CN 4-C1 0 m.p. H CH ₃ H H 3-C1 4-CN 4-CF ₃ 0 m.p. H CH ₃ H H 3-C1 4-CN 4-CF ₃ 0 m.p. H CH ₃ H H 3-C1 H CH ₃ H H B-CN 4-CN 6-CF ₃ 0 m.p. H CH ₃ H H 4-C1 H C-CN 6-CC 0 m.p.	H CH ₃ H H H 4-C1 3,4-C1 ₂ 0 Paste H CH ₃ H H H 4-C1 3-CF ₃ 0 Paste H CH ₃ H H H 4-C1 3-CF ₃ 0 Paste H CH ₃ H H 3-C1 4-CN 4-CI 0 M.P. H CH ₃ H H 3-C1 4-CN 4-CF ₃ 0 M.P. H CH ₃ H H 3-C1 4-CN 4-CF ₃ 0 M.P. H CH ₃ H H 4-C1 H 2-C1 0 Paste H CH ₃ H H 4-C1 H 2-C1 0 Paste H CH ₃ H H 4-C1 H 3-C1 0 Paste	H CH ₃ H H H 4-Cl 3,4-Cl ₂ O Paste H CH ₃ H H H 4-Cl 3-CF ₃ O Paste H CH ₃ H H H 4-Cl 3-CF ₃ O m.p. H CH ₃ H H 3-Cl 4-CN 4-CN 4-CF ₃ O m.p. H CH ₃ H H 3-Cl 4-CN 4-CF ₃ O m.p. H CH ₃ H H 3-Cl 4-CN 4-CF ₃ O m.p. H CH ₃ H H 4-Cl H 2-Cl O Paste H CH ₃ H H 4-Cl H 3-Cl O Paste H CH ₃ H H 4-Cl H 3-Cl O Paste H CH ₃ H H 4-Cl H 3-Cl O Paste H CH ₃ H H 4-Cl H 3-Cl O Paste H CH ₃ H H 4-Cl H 3-Cl O Paste	H CH ₃ H H H 4-CI 3,4-Cl ₂ O Paste H CH ₃ H H H 4-CI 3-CF ₃ O Paste H CH ₃ H H H A-CI 4-OCF ₃ O Paste H CH ₃ H H 3-CI 4-CN 4-CI 0 Paste H CH ₃ H H 3-CI 4-CN 4-CI 0 Paste H CH ₃ H H 4-CI H 2-CI O Paste H CH ₃ H H 4-CI H A-CI O Paste H CH ₃ H H 4-CI H A-CI O Paste H CH ₃ H H 4-CI H A-CI O Paste H CH ₃ H H 4-CI H A-CI O Paste H CH ₃ H H 4-CI H A-CI O Paste H CH ₃ H H 4-CI H A-CI O Paste H CH ₃ H H 4-CI H H 4-CI O Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H 4-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H CH ₃ H H H A-CI O D Paste H H A-CI O D P Paste H A-CI O D P P P P P P P P P P P P P P P P P P	H CH ₃ H H H H 4-C1 3.4-C1 ₂ O Paste H CH ₃ H H H H H H-C1 3-CF ₃ O Paste H CH ₃ H H H H H H-C1 4-C1 3-CF ₃ O M.p. H CH ₃ H H 3-C1 4-CN 4-CF ₃ O M.p. H CH ₃ H H 3-C1 4-CN 4-CF ₃ O M.p. H CH ₃ H H 4-C1 H 2-C1 O Paste H CH ₃ H H 4-C1 H 4-CN H 4-CF ₃ O M.p. H CH ₃ H H 4-C1 H 4-C1 O Paste H CH ₃ H H 4-C1 H 4-C1 O Paste H CH ₃ H H 4-C1 H 4-C1 O Paste H CH ₃ H H 4-C1 H 4-CT O Paste H CH ₃ H H 4-C1 H H 4-C1 O M.p. H A-CT O M.p. H CH ₃ H H 4-C1 O M.p. H A-CT O M.p. H A-CT O M.p.	H CH ₃ H H H 4-C1 3.4-C1 ₂ O Paste H CH ₃ H H H 4-C1 3-CF ₃ O Paste H CH ₃ H H H 4-C1 3-CF ₃ O Paste H CH ₃ H H A-C1 4-CN 4-CI 0 M.P. H CH ₃ H H 3-C1 4-CN 4-CF ₃ O M.P. H CH ₃ H H 4-C1 H CH ₃ H H 4-C1 H CH ₃ H H 4-C1 H A-CN 4-CF ₃ O M.P. H CH ₃ H H 4-C1 H A-C1 O Paste H CH ₃ H H 4-C1 H A-C1 O Paste H CH ₃ H H 4-C1 H A-C1 O Paste H CH ₃ H H 4-C1 H A-C1 O Paste H CH ₃ H H 4-C1 H A-CCF ₃ O M.P. H CH ₃ H H 4-C1 H A-CCF ₃ O Paste H CH ₃ H H 4-C1 H A-CCF ₃ O Paste H CH ₃ H H 4-C1 H A-CCF ₃ O Paste H CH ₃ H H 4-C1 H A-CCF ₃ O Paste	H CH ₃ H H H 4-C1 3.4-C1 ₂ O Paste H CH ₃ H H 4-C1 3.6F ₃ O Paste H CH ₃ H H A-C1 3-CF ₃ O Paste H CH ₃ H H A-C1 4-CC G M.p. H H CH ₃ H H A-C1 4-CC G M.p. H CH ₃ H H 3-C1 4-CN 4-CF G M.p. H CH ₃ H H A-C1 H CH ₃ H H A-C1 H A-CN A-CCF ₃ O M.p. H CH ₃ H H A-C1 H A-C1 D Paste H CH ₃ H H A-C1 H A-C1 D Paste H CH ₃ H H A-C1 H A-C1 D Paste H CH ₃ H H A-C1 H A-CCF G M.p.	H CH ₃ H H H 4-C1 3.4-C1 ₂ O Paste H CH ₃ H H H 4-C1 3-CP ₃ O m.p. H H CH ₃ H H H 4-C1 3-CP ₃ O m.p. H H CH ₃ H H H 4-C1 4-CN 4-CI 0 m.p. H CH ₃ H H 3-C1 4-CN 4-CP ₃ O m.p. H CH ₃ H H 3-C1 H 4-CN 4-CP ₃ O m.p. H CH ₃ H H 4-C1 H 4-CN 4-CP ₃ O m.p. H CH ₃ H H 4-C1 H 4-CN H 4-CP ₃ O m.p. H CH ₃ H H 4-C1 H 4-C1 H 4-CP ₃ O m.p. H A-CP ₃ O m.p. H A-CP ₃ D M.p. D Paste H CH ₃ D M.p. H A-CP ₃ D M.p. D Paste H CH ₃ D M.p. D Paste D D Paste D D D D D D D D D D D D D D D D D D D

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5		m.p. 188°C	m.p. 176°C	m.p. 170°C	Paste	m.p. 185°C E-form	m.p. 95°C Z-form	Paste	m.p. 113°C	Paste	m.p. 76°C	m.p. 142°C	m.p. 149°C	m.p. 141°C
10		0	0	0	0	0	0	0	0	0	0	0	0	0
70														
15		4-C1	4-CF ₃	4-0CF ₃	4-C1	4-0CF ₃	4-OCF	4-C1	4-0CF3	4-C1	4-0CF3	н	4-C1	4-CF3
20														
25	(cont'd)	Ħ	н	н	4-C1	4-C1	4-C1	4-CN	4-CN	2-C1	2-C1	3-C1	3-C1	3-C1
30	Table 3								-	r-4	·	H		T.
35		Ħ	H	Ħ	Ħ	Ħ	Ħ	×	Ħ	2-C1	2-C1	3-C1	3-C1	3-C1
		Ħ	#	Ħ	Ħ	Ħ	Ħ	н	н	Ħ	Ħ	Ħ	н	н
40		НО	НО	НО	НО	НО	Ю	НО	ЮН	ОН	ОН	НО	НО	ОН
		Ħ	æ	H	Ħ	I	Ħ	Ħ	ж	Ħ	Ħ	H	н	н
45		ж	Ħ	æ	Ħ	н	н	H	Ħ	н	×	I	Ħ	н
		A331	A332	A333	A334	A335	A336	A337	A338	A339	A340	A341	A342	A343
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5		m.p. 146°C	m.p. 81°C	m.p. 59°C	Paste	Paste	m.p. 72°C	m.p. 100°C	m.p. 88°C	m.p. 168°C	m.p. 180°C	m.p. 184°C	m.p. 182°C	m.p. 139°C	m.p. 142°C	m.p. 178°C
10		0	0	0	0	0	0	0	0	0	0	0	0	0	0	Ø
15		4-0CF ₃	3-61	4-C1	4-CF3	4-0CF ₃	4-0CF ₃	3,4-C1 ₂	4-C1	4-0CF ₃	4-C1	4-CF3	4-0CF ₃	4-C1	4-0CF ₃	4-0CF ₃
25	(cont'd)	3-C1	4-C1	4-C1	4-C1	4-C1	2,4-C1 ₂	2,4-C1 ₂	4 - 표	4-F	4-CH ₃	4-CH ₃	4-CH ₃	4-0CH ₃	4-0CH ₃	4-CH ₃
30 35	Table 3	3-C1	4-C1	4-C1	4-C1	4-C1	2,4-Cl ₂	2,4-Cl ₂	4-F	4-F	4-CH ₃	4-CH ₃	4-CH ₃	4-0CH ₃	4-0CH ₃	4-CH ₃
	ļ	#	Ħ	н	н		Ħ	н	H	Ħ	=	×	Ħ	Ħ	Ħ	Н
40		НО	ОН	НО	НО	HO HO	ОН	HO HO	НО	НО	НО	Он	НО	НО	ОН	ОН
45		H	Ħ	Ħ	æ	=	#	H	#	Ħ	Ħ	=	Ħ	Ħ	Ħ	н
45		H	×	H	H	×	×	×	H	×	×	Ħ	Ħ	×	H	Н
50		A344	A345	A346	A347	A348	A349	A350	A351	A352	A353	A354	A355	A356	A357	A358
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5		m.p. 167°C	164°C	150°C	132°C	118°C	197°C	183°C	192°C	185°C	181°C	155°C	209°C	193°C	J.94I	184°C
			m.p.	m.p.	m.p.	m.p.	m.p.	a.₽.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	g.E
10		0	0	0	0	လ	0	0	0	0	0	0	0	0	0	0
	i															
15		4-0CF3	4-C1	4-CF3	4-CF3	4-0CF3	3-C1	4-C1	$3-CF_3$	4-CF3	4-0CF3	4-C1	4-CF ₃	4-0CF3	4-C1	4-0CF ₃
20																
	(cont'd)	н	н	Ħ	н	H	н	н	H	н	#	H	æ	н	H	Н
25	Table 3 (c	4-N(CH ₃) ₂	-	_			۵						_			
30	Та	4	==	<u> </u>	=	=	=	=	=	<u> </u>	<u> </u>	=	=	=	=	=
		н	н	н	Ħ	#	H	#	#	н	H	Ħ	#	×	=	н
35																
40		НО	СН3	снз	CH ₃	CH ₃	оснз	och ₃	och ₃	och ₃	осн3	0C3H7-i	oc3H7-i	oc3H7-i	OC4H9-i	oc4H9-i
		н	н	ж	=	#	Ħ	Ħ	#	Ħ	Ħ	Ħ	#	Ħ	×	#
45		H	Ħ	Ħ	H	H	H	н	H	H	Ħ	н	H	н	H	н
50		A359	A360	A361	A362	A363	A364	A365	A366	A367	A368	A369	A370	A371	A372	A373
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	45		35 40		30	25	20	15	10	5	
				-	Table 3 ((cont'd)					
A374	#	Н	ососнз	æ	н	æ	4,	4-0CF ₃	0	m.p. 182°C	Ų
A375	Ħ	Ħ	00-00-0	Ħ	Ħ	Ħ	4	4-0CF3	0	m.p. 168°C	ပ
A376	Ħ	æ	но	CH ₃	ж	Ħ	4	4-C1	0	m.p. 115°C	ပ
A377	H	H	НО	CH ₃	I	Ħ	4,	4-OCF ₃	0	m.p. 130°C	ပ
A378	Ħ	×	II	O _{II}	Ħ	Ħ	4-	CF ₃	0	m.p. 150°C	ပ
A379	×	Ħ	II	0=	н	н	4	4-0CF ₃	0	m.p. 132°C	υ
A380	×	Ħ	æ	×	н	2,4-F ₂	4-	4-C1	0	m.p. 199°C	ပ
A381	Ħ	Ħ	Ħ	Ħ	= .	2,4-F ₂	4-	4-0CF ₃	0	m.p. 173°C	ပ္
A382	Ħ	Ħ	Ħ	æ	н	3,4-F2	4-	4-C1	0	m.p. 203°C	ပွ
A383	Ħ	×	Ħ	н	н	3,4-F2	4-	4-0CF ₃	0	m.p. 189°C	ပ
A384	Ħ	エ	н	==	H	3,4-F2	4-	$4-SCF_3$	0	m.p. 207°C	ပွ
A385	Ħ	H	Ħ	×	н	3,4-F2	4-	4-SOCF ₃	0	m.p. 188°C	ပ
A386	н	Ħ	Ħ	ж	н	3,4-F2	-4	$4-SO_2CF_3$	0	m.p. 194°C	ပ
A387	Ħ	Ħ	н	Ħ	Н	3,5-F2	4	4-C1	0	m.p. 205°C	ပ
A388	Ħ	Ħ	r:	Ħ	н	3,5-F2	4-	4-Br	0	m.p. 201°C	ပ

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5		m.p. 196°C	m.p. 186°C	m.p. 208°C	m.p. 215°C	m.p. 186°C	m.p. 187°C	m.p. 196°C	m.p. 170°C	m.p. 193°C	m.p. 209°C	Glass-like amorphous	m.p. 168°C	m.p. 204°C
10		0	0	0	0	0	0	0	0	0	0	0	0	0
15		4-0CF ₃	4-0C ₄ H _{9-t}	4-C1	4-CF ₃	4-0CF ₃	$4-0$ CF $_2$ CHF $_2$	4-0CF ₃	4-ocF ₃	4-0CF3	$4-0$ CF $_3$	4-SOCF ₃	4-0CF3	4-0CF ₃
20														
25	3 (cont'd)	3,5-F2	4-C1	3,4-C1 ₂	3,4-C1 ₂	3,4-C1 ₂	3,4-C1 ₂	4-CF3	\$ P	4-NHCHO	4-NHCONHC ₂ H ₅	4-NO ₂	4-0CF3	4-0CF ₃
30	Table	H	н	Н	H	H	æ	н	н	#	Ħ	# #	H	Ħ
35		Ħ	#	×		=	Ħ	Ħ	=	н	н	Ħ	н	æ
40	}	Ħ	н	æ	н	н	н	Ħ	¤	н	н	н	н	H
		H	H	Ħ	Ħ	H	#	×	Ħ	Ħ	ж	H	Ħ	н
45		Ħ	<u> </u>	=	H	<u> </u>	<u> </u>	<u> </u>	 =	 #	H	H	<u> </u>	н
	Ī	A389	A390	A391	A392	A393	A394	A395	A396	A397	A398	A399	A400	A401
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m.p. 169°C m.p. 214°C m.p. 194°C m.p. 166°C m.p. 177°C m.p. 228°C m.p. 234°C m.p. 186°C m.p. 221°C m.p. 224°C m.p. 165°C m.p. 237°C m.p. 255°C m.p. 207°C m.p. 173°C 5 0 0 0 0 0 0 0 0 0 0 0 0 10 2,3,4,5-Cl₄ 2,3,4-Cl₃ $4-50_2$ CF $_3$ 4-0CF₂Br 4-SOCF3 15 4-SOCF3 4-SCF₃ 4-0CF₃ 4-SCF3 4-0CF3 4-0CF3 4-C1 20 $4-50_2N(CH_3)_2$ $4-SO_2N(CH_3)_2$ Table 3 (cont'd) 4-OCH2CF3 4-SOCHF3 4-SCHF3 4-CN 4-CN 25 4-CN 4-CN 4-CN 4-CN 4-CN 4-CN 4-CN 4-CN 30 3-C1 3-C1 3-C1 3-C1 4-F 3-F 35 I H H H Ξ 二 王 Ξ H Ξ H H 40 I Ξ I Ξ Ξ Ξ I I Ξ 二 I Ξ Ξ H Ħ H I I H H Ħ I Ħ I Ξ H ${\tt H}$ H 45 I Ħ Ή Ξ Ξ A415 A416 A410 A412 A414 A408 A413 A405 A406 A409 A411 A403 A404 A407

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	CH ₃ 0 m.p. 218°C	3-C1-4-OCF ₂ CHF ₃ 0 m.p. 211°C	4-0-COCH ₃ 0 m.p. 177°C	4-0SO ₂ CF ₃ 0 m.p. 199°C	CF ₃ 0 m.p. 157°C Z-form		3 O m.p. 199°C	F ₃ 0 m.p. 188°C	F ₃ 0 m.p. 197°C	F ₃ 0 m.p. 180°C	O m.p. 209°C	F ₃ 0 m.p. 164°C	F ₃ O m.p. 189°C	CF ₃ O m.p. 207°C	2 ^{CF} 3 O m.p. 205°C
	4-COCH ₃	3-C1	4-0-	4-08	4-SOCF3		4-CF3	4-0CF ₃	4-SCF ₃	4-0CF3	4-C1	4-0CF3	4-SCF3	4-SOCF3	4-SO ₂ CF ₃
o (t,q)						į	Ş	Ç	ÇN	CN	-CN	-CN	-CS	-CN	-CN
3 (cont'd)	4-CN	4-CN	4-CN	4-CN	4-CN	;	2-C1-4-CN	2-C1-4-CN	2-C1-4-CN	3-C1-4-CN	2-CH ₃ -4-CN	2-CH ₃ -4-CN	2-CH ₃ -4-CN	2-CH ₃ -4-CN	2-CH ₃ -4-CN
Table	3-c1	3-C1	3-C1	3-C1	3-C1	;	3-C1	3-C1	3-C1	3-C1	3-C1	3-C1	3-01	3-C1	3-C1
35	Ħ	н	Ħ	Ħ	Ħ	:	E.	H	Ħ	Ħ	Ħ	Ħ	Ħ	×	F
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	#	H	#	王	H	:	E	H	н	н	Ħ	н	Ħ.	Ħ	Ħ
45	#	Ħ	×	Ħ			۲	Ħ	H	Ħ	Ħ	Ħ	Ħ	H	#
50	A417	A418	A419	A420	A421	C C 7 K	A422	A423	A424	A425	A426	A427	A428	A429	A430

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50	45		40	35	30	20	15	10		5
					Table	3 (cont'd)				
A431	H	H	Н	н	3-C1	3-CH ₃ -4-CN	4-C1	0	m.p.	m.p. 209°C
A432	ж	×	Ħ	H	3-C1	3-CH ₃ -4-CN	4-0CF ₃	0	m.p.	m.p. 199°C
A433	Ħ	Ħ	Ħ	æ	3-C1	3-CH ₃ -4-CN	4-SCF ₃	0	m.p.	213°C
A434	H	H	ж	=	3-C1	3-CH ₃ -4-CN	4-SOCF ₃	0	m.p.	180°C
A435	Ħ	Ħ	Œ	I	3-C1	3-CH ₃ -4-CN	4-SO ₂ CF ₃	0	m.p.	152°C
A436	Ħ	н	Ħ	Ħ	3-C1	3,4-(CN) ₂	4-0CF ₃	0	m.p.	211°C
A437	Ħ	н	Ħ	H	3-C1	4-C00CH ₃	4-0CF ₃	0	m.p.	J.091
A438	н	н	Ħ	н	3-C1	4-0H	4-0CF ₃	0	m.p.	193°C
A439		エ	Ħ	щ	3-C1	4-0CHF ₂	4-CF ₃	0	m.p.	181°C
A440	Ħ	H	Ħ	ж	3-C1	4-0CHF ₂	4-0CF ₃	0	m.p.	170°C
A441	=	Ħ	Ħ	×	3-C1	4-OCHF ₂	4-SCF3	0	m.p.	193°C
A442	I	Ħ	H	Ħ	3-C1	4-050 ₂ CH ₃	4-0CF ₃	0	m.p.	195°C
A443	#	н	Н	Ħ	3-C1	4-0SO ₂ CF ₃	4-0CF3	0	m.p.	173°C
A444	H	Ħ	Ħ	Ħ	3-Br	4-CN	2,3,4-Cl ₃	0	m.p.	247°C
A445	H	Ħ	н	Ħ	3-Br	4-CN	2,3,4,5-Cl ₄	0	m.p.	250°C
	_							_		

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5		m.p. 169°C	228°C	177°C	172°C	215°C	210°C	225°C	211°C	J.96I	197°C	190°C	J.691	224°C	
		ď.m	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.	g.e	m.p.	
10		0	0	0	0	0	0	0	0	0	0	0	0	0	
15		4-C ₆ H ₁₃ -n	$3-\text{Cl}-4-\text{OCF}_2\text{CHF}_2$	4-0CF ₃	$4-0$ CF $_2$ Br	4-SCF ₃	4-SOCF ₃	$4-SO_2CF_3$	4-0CF ₃	4-0CF ₃	4-CF ₃	4-0CF ₃	4-0CF ₃	4-ocr ₃	
20									-						\dashv
25	(cont'd)	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4~CN	4-CN	4-CN	4-CN	
30	Table 3	3-Br	3-Br	3-CHF ₂	3-CF ₃	3-CF3	3-CF3	3-CF3	$4-CF_3$	3-F-5-CF3	$3-CH_2OCH_3$	3-CH ₂ 0CH ₃	3-04	3-ОН	
35															7
		Ħ	耳	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	×	×	×	Ħ	Ħ	
40		E	=	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	Ħ	æ	н	ж	н	
		Ħ	Ħ	Ħ	Ħ	H	H	H	-		174	lyel .	244	Park .	
45	ŀ		74		-				Ξ_	<u> </u>	<u> </u>	Ξ	<u>#</u>	<u> </u>	\dashv
		=======================================	Ħ	Ħ	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ		Ħ	#	╛
50		A446	A447	A448	A449	A450	A451	A452	A453	A454	A455	A456	A457	A458	

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50	45		40		35	30	25	20	15	10		5	
					Га	Table 3	(cont'd)						
1459	Ξ	н	×	=	3-och ₂ ch=ch ₂	:CH ₂	4-CN	4-0CF3		0	m.p.	m.p. 160°C	
1460	×	Ħ	Ħ	田	3-0CHF ₂		4-CN	4-0CF3		0	m.p.	188°C	
1461	H	Ħ	н	Ħ	3-OCHF ₂	,-	4-CN	4-SCF3		0	m.p.	204°C	
1462	Ħ	Ħ	н	Ħ	3-OCHF ₂	<u> </u>	4-CN	4-SOCF3		0	g.p.	195°C	
4463	Ħ	Ħ	ш	H	3-OCHF ₂		4-CN	4-SO2CF3		0	m.p.	206°C	
4464	Ħ	Ħ	H	H	3-ocF3		4-CN	4-CF3		0	m.p.	159°C	
4465	×	Ħ	H	н	3-0CF ₃		4-CN	4-0CF3		0	m.p.	171°C	
4466	ж	×	€ H	Ħ	3-0CF3		4-CN	4-SCF3		0	m.p.	202°C	
A467	H	×	H	н	3-0CF3		4-CN	4-SOCF3		0	m.p.	200°C	
A468	Ħ	н	H	H	3-0CH2COCH3	ж3	4 CN	4-0CF3		0	m.p.	198°C	
A469	Ħ	×	H	H	3-0CH2CO2C2H5	C2H5	4-CN	4-0CF3		0	m.p.	172°C	
A470	Ħ	Ħ	н	н	3-0-COC3H5-c	12-c	4-CN	4-0CF3		0	m.p.	200°C	
A471	Ħ	Ħ	Ħ	Ħ	3-0-CON(CH ₃) ₂	H ₃) ₂	4-CN	4-0CF3		0	m.p.	197°C	
A472	н	Ħ	Ħ	н	3-0-C0 ₂ CH ₃	13	4-CN	4-0CF3		0	m.p.	197°C	
A473	Ħ	Ħ	Ħ	н	3-0S0 ₂ CH ₃	~	4-CN	4-0CF3		0	m.p.	180°C	

cont'd -

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5		m.p. 201°C	m.p. 248°C	m.p. 207°C	m.p. 231°C	m.p. 152°C Z-form	m.p. 165°C	m.p. 184°C	m.p. 178°C	m.p. 148°C	m.p. 214°C	m.p. > 214°C	Hydro- chloride
10		0	0	0	•	0	0	0	ß	w	0	0	
15		4-SC ₃ F ₇ -n	4 C1	4-SOC ₃ F ₇ -n	4-SO ₂ C ₃ F ₇ -n	4-0CF3	4-C1	4-0CF ₃	4-C1	4-CH ₃	4-0CF ₃	4-ocF ₃	
20											-4	-4	
25	Table 3 (cont'd)	4-CN	4 - CN	4-CN	4-CN	4-CN	4-CN	4-CH ₃	4-NO ₂	4-CN	3-CH=CH-CH=N-4	3-CH=CH-CH=N-4	
30	Table	3-CF3	3-CF3	3-CF3	3-CF3	3-CF ₃	3-CF3	3-CF ₃	4-F	유 표	Ħ	æ	
35	·	3	<u></u>	<u></u>			(7)	(*)	4				
		H	Ħ	Ħ	Ħ	Ħ	H	×	Ħ	Ħ	Ħ	Ħ	
40		н	H	Ħ	н	Ħ	н	Н	ж	Ħ	H	Ħ	
		H	H	н	н	н	н	н	H	н	Ħ	Ħ	
45		#	=	#	н	н	н	ж	Ħ	H	н	Œ	
50		A474	A475	A476	A477	A478	A479	A480	A481	A482	A483	A484	

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Table 3 (cont'd)

}	×	Ħ	Ħ	н	3-CH=CH-CH=N-4 4-ClF	4-ClF	0	0 m.p. 210°C
A486 H	Ħ	Ħ	Ħ	3-CF3	4-CN	4	0	0 m.p. 221°C
					-	c1\\c1		

In the table, -c denotes an alicyclic compound and m.p. denotes melting point.

Table 4 shows 1-NMR data of the compounds having physical properties as paste listed in Table 3.

Table 4

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No.	¹ H-NMR(CDC1 ₃ /TMS, & value, ppm.)
A055	3.88+4.23(s, 2H), 6.52+6.54(t, 1H),
	6.95+7.77(m, 13H), 8.73+8.98+9.20+9.39(s, 2H).
!	(Mixture of E- and Z-forms)
A307	2.70(s, 3H), 3,87(s, 2H), 6.90-7.60(m, 13H),
	8.23(bs, lH).
A309	2.23(s, 3H), 3.17(s, 3H), 4.20(s, 2H),
	6.50-7.83(m, 15H).
A311	2.73+3.20(s, 3H), 3.70(s, 3H),
	3.90+4.23(s, 2H), 6.60-8.00(m, 14H),
	8.17(bs, lH). (Mixture of E- and Z-forms)
A316	2.60+3.20(s, 3H), 3.87+4.20(s, 2H),
	6.27-8.27(m, 13H). (Mixture of E- and Z-forms)
A317	2.73+3.23(s, 3H), 3.90+4.23(s, 2H),
	6.73-8.13(m, 13H), 8.40(bs, 1H). (Mixture of
	E- and Z-forms)
A322	3.23(s, 3H), 4.20(s, 2H), 6.67-8.43(m, 13H),
	8.77(bs, lH).
A323	2.73+3.23(s, 3H), 3.87+4.20(s, 2H),
	6.80-8.00(m, 14H). (Mixture of E- and Z-forms)
	A307 A309 A311 A316 A317

- cont'd -

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Table 4 (cont'd)

5	A326	2.73+3.20(s, 3H), 3.87+4.20(s, 2H),
		6.43-7.93(m, 13H), 8.00(bs, 1H).
		(Mixture of E- and Z-forms)
10	A327	2.73(s, 3H), 3.87(s, 1H), 6.73-7.90(m, 13H),
		8.30(bs, lH).
15	A328	2.23+2.33(s, 3H), 2.70+3.17(s, 3H),
		3.87+4.17(s, 2H), 6.43-7.90(m, 13H).
		(Mixture of E- and Z-forms)
20	A329	2.30(s, 2H), 3.17(s,3H), 4.17(s, 2H),
		6.83-8.30(m, 14H).
25 .	A334	6.29(s, 1H), 7.65-7.92(m, 13H), 9.14(bs, 1H),
20		10.70(bs, lH), (DMSO-d ₆)
	A337	3.88(bs, 1H), 3.87(s, 1H), 6.91-7.55(m, 13H),
30		7.73(s, 1H), 8.13(bs, 1H).
	A339	3.72(bs, 1H), 6.08(s, 1H), 6.77-7.68(m, 12H),
		8.17(bs, 1H), 10.58(bs, 1H).
35	A347	3.75(bs, lH), 5.60(d, lH), 6.94-7.61(m, 12H),
		8.18(s, 1H), 10.80(s, 1H).
40	A348	3.75(bs, lH), 5.31(d, lH), 6.92-7.65(m, 12H),
		8.13(s, lH), 10.75(s, lH).
	A399	4.31(s, 2H), 7.31-8.14(m, 13H), 8.63(s, 1H),
45		9.15(s, 1H).

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$ \begin{array}{c}     W \\                               $
( ² ) -N
C-N(R
(R ¹ ) -(
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N-O

Table 5 (Wherein each of R  1  and R  4  is a hydrogen atom.)

(I") or (I")

No.	No. R ² R ³		R5	×	А	Z	3	Physical properties	al
B001	н	Н	н	Н	Н	4-C1	0	m.p. 211°C	11°C
B002	Ħ	H	н	Ħ	耳	4-0CF ₃	0	m.p. 194°C	14°C
B003		Ħ	н	æ	4-C1	4-0CF ₃	0	m.p. 209°C	2.60
B004		н	H	ĸ	4-C1	4-OCF2CHF2	0	m.p. 202°C	2°C
B005	Ħ	Ħ	Ħ	н	4-C1	$4-0$ $CF_3$ $0$	0	m.p. 215°C	2,61
B006	н	н	н	н	4-CN	4-0CF ₃	0	m.p. 204°C	)4°C

- cont'd -

	45	45	40	35	30	25	20	15	10		5
					Table 5	(cont'd)					
B007	#	н	Ħ		Н	4-NHCOCH ₃		4-0CF ₃	0	m.p.	206°C
B008	H	Ħ	н		F	4-NO ₂		4-0CF3	0	m.p.	188°C
B009	×	Ħ	Ħ		н	4-0CHF ₂		4-0CF ₃	0	m.p.	202°C
B010	Ħ	×	Ħ		3-F	4-C1		4-0CF ₃	0	m.p.	203°C
B011	æ	Ħ	<b>=</b>		4 - F	4-C1		4-0CF3	0	m.p.	207°C
B012	<b></b>	н	щ		3-C1	4-C1		4-CF ₃	0	m.p.	189 °C
B013	ж	н	Ħ		3-C1	4-C1		4-0CF3	0	m.p.	176°C
B014	Ħ	H	Ħ		3-C1	4-CN		4-CF ₃	0	m.p.	198°C
B015	×	н	н		3-C1	4 - CN		4-0CF ₃	0	m.p.	193°C
B016	<b>=</b>	Ħ	H		3-C1	4-CN		4-OCF2CHF3	0	m.p.	164°C
B017	×	н	Ħ		3-C1	4-CN		4-0C4H9-t	0	m.p.	186°C
B018	н	Ħ	Ħ		3-C1	4-CN		4-SCF3	0	m.p.	177°C
B019	H	×	н		3-C1	4-CN		4-SOCF ₃	0	m.p.	178°C
B020	н	Ħ	Ħ		3-C1	4-CN		$4-SO_2CF_3$	0	m.p.	170°C
B021	н	Ħ	H		4-C1	4-C1		4-CF3	0	m.p.	195°C

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	45		40	35	30	25	20	15	10	5	
					Table 5	(cont'd)					
	н	×	Н		4-C]	4-C1		4-0CF ₃	0	m.p.	165°C
_	Ħ	Ħ	Ħ	·	4-C1	4-CN		4-0CF ₃	0	m.p.	210°C
	<b>=</b>	Ħ	н	<del></del>	3-Br	4-CN		4-0CF ₃	0	m.p.	187°C
	#	н	耳		3-CF3	4 -CN		4-0CF ₃	0	m.p.	165°C
	Ħ	Ħ	Ħ		3-CF3	4-CN		4-SCF ₃	0	m.p.	164°C
	Ħ	н	H		4-OCHF2	н	····	4-C1	0	m.p.	192°C
	Ħ	н	Ħ	·· <del> ·· ·</del> · ·	4-OCHF2	Ħ	•	4-0CF ₃	0	m.p.	217°C
	Ħ	н	<b>¤</b>		4-0CHF2	н		4-SCF ₃	0	m.p.	209°C
	H	Ħ	<b>=</b>		3-0	4 -CN		4-0CF ₃	0	m.p.	164°C
	E	н	×		Ħ	4-C1		4-0CF ₃	Ø	m.p.	171°C
	×	Ħ	ж		3-C1	4-CN		4-0CF ₃	ß	m.p.	149°C
	Ħ	н	Ħ		4-C1	4-CN		4-0CF ₃	Ø	m.p.	195°C
	H	н	Ħ		3-CF3	4 -CN		4-0CF ₃	ß	m.p.	209°C
	E	Ħ	соснз		3-C1	4-CN		4-0CF ₃	0	m.p.	178°C
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50	45	45	40	35	30	25	20	15	10		5
					Table 5	(cont'd)					
B036	H	н	© 8		3-C1	4-CN		4-0CF ₃	0	m.p.	m.p. 221°C
B037	#	н	CONHC ₂ H ₅		3-C1	4-CN		4-0CF ₃	0	m.p.	201°C
B038	Ħ	ЮН	Ħ,		Ħ	Ħ	<del></del>	4-CF ₃	0	m.p.	200°C
B039	Ħ	НО	н		ж	=		4-0CF3	0	m.p.	190°C
B040	H	осн3	н	<del></del>	Ħ	Ħ		4-C1	0	m.p.	195°C
B041	Ħ	осн3	Ħ		н	H		4-0CF3	0	m.p.	183°C
B042	H	осн3	Ħ		н	н		4-0CF ₃	0	m.p.	186°C
B043	СНЗ	Ħ	æ		3-C1	4-CN		4-0CF ₃	0	m.p.	156°C
B044	н	Ħ	Ħ		н	4-F		4-CF3	0	m.p.	211°C
B045	H	Ħ	Ħ		Ħ	4-F		4-0CF ₃	0	m.p.	209°C
B046	Ħ	н	Ħ		H	2,4-F ₂		4-C1	0	m.p.	229°C
B047	Ħ	Ħ	ж		H	2,4-F2		4-0CF ₃	0	m.p.	212°C
B048	III.	×	<b>H</b>		н	3,4-F2		ប	0	m.p.	201°C
B049	<b>=</b>	Ħ	Ħ		н	3,4-F2		4-0CF ₃	0	m.p.	170°C
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5	0 m.p. 163°C	o m.p. 163°C	0 m.p. 182°C	O m.p. 185°C	0 m.p. 194°C	0 m.p. 233°C	0 m.p. 227°C	0 m.p. 201°C	0 m.p. 218°C	0 m.p. 201°C	0 m.p. 215°C	0 m.p. 186°C	0 m.p. 176°C	0 m.p. 153°C	0 m.p. 188°C
10								· · · · · · · · · · · · · · · · · · ·			-				
15	4-SCF ₃	4-SOCF3	4-SO ₂ CF ₃	4-C1	4-Br	4-C1	4-CF3	4-0CF3	4-CF3	4-0CF3	4-0CF ₃	4-CF3	4-0CF3	4-OCF ₂ CHF ₂	4-0CF ₃
20															
(cont 'd)	3,4-F2	3,4-F2	3,4-F2	3,5-F2	3,5-F2	4-Br	4-Br	4-Br	4-CH ₃	4-CH ₃	4-CF3	3-CN	3-CN	3-CN	4-NH ₂
Table 5	н	F	H	Ħ	н	H	Ħ	Ħ	Н	#	H	н	Ħ	Н	н
35															
10	н	н	Œ	Ħ	Ħ	E	<b>E</b>	エ	H	#	Ħ	エ	Ħ	Ħ	H
	Ħ	н	Ħ	·#	Ħ	Ħ	Ħ	H	ж	#	Ħ	#	Ħ	#	<b>=</b>
5	ж	Ħ	Ħ	Ħ	æ	н	Ħ	ж	Ħ	===	Ħ	æ	<b></b>	æ	×
:o	B050	B051	B052	B053	B054	B055	B056	B057	B058	B059	во60	B061	B062	B063	B064

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50	45		35 40	30	20	10		5	
				Table 5	Table 5 (cont'd)				
B065	H	×	ж	н	2-NO ₂	4-0CF ₃	0	m.p.	m.p. 197°C
B066	H	Ħ	ж	<b>H</b>	4-NO ₂	4-SCF ₃	0	m.p.	188°C
B067	н	н	Ħ	エ	4-0CF ₃	4-0CF3	0	m.p.	206°C
B068	×	H	Ħ	<b>H</b>	4-0CF ₃	4-SCF ₃	0	m.p.	181°C
B069	н	H	×	ш	4-SCHF2	4-0CF ₃	0	m.p.	195°C
B070	Ħ	×	н	 F.	4-CN	4-0CF ₃	0	m.p.	189°C
B071	Ħ	H	coch ₃	3-5	4-CN	4-0CF ₃	0	m.p.	193°C
B072	Ħ	н	COC4H9-t	3-F	4-CN	4-0CF ₃	0	m.p.	218°C
B073	Ħ	Ħ	#	3-F	4-CN	4-0CF ₂ Br	0	m.p.	201°C
B074	Ħ	н	<b>=</b>	3-F	4-CN	4-SCF ₃	0	m.p.	189°C
B075	Ħ	н	н	 연 -	4-CN	4-SOCF ₃	0	m.p.	166°C
B076	Ħ	н	r.	3-F	$4-SO_2N(CH_3)_2$	4-C1	0	m.p.	216°C
B077	Ħ	×	Ħ	3 - F	$4-SO_2N(CH_3)_2$	4-0CF3	0	m.p.	214°C
B078	H	×	×	4 - 된	4-F	4-0CF ₃	0	m.p.	174°C
B079	Ħ	Ħ	Ħ	4 - F	4-CN	4-0CF ₃	0	m.p.	194°C
							_		

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50	45		35 40	30	20 25	15	10		5
				Table 5 (cont'd)	(cont'd)				
B080	н	Ħ	ж	4-F	4 - CN	4-SCF ₃	0	ļ	m.p. 211°C
B081	ж	Ħ	н	4-F	4-CN	4-SOCF3	0	ď.E	, 198°C
B082	H	#	×	4-F	4 -CN	4-SO ₂ CF ₃	0		m.p. 177°C
B083	æ	#	Ħ	3-C1	4 ()	4-0CF ₃	0		m.p. 207°C
B084	Ħ	н	н	3-C1	4 -CN	4-1	0		m.p. 218°C
B085	æ	· H	COC ₂ H ₅	3-C1	4 -CN	4-0CF ₃	0	m.p.	. 202°C
B086	Œ	Ħ	COC ₃ H ₇ -n	3-01	4-CN	4-0CF3	0	m.p.	. 142°C
B087	Ħ	×	coc ₃ H ₅ -c	3-01	4-CN	4-0CF3	0	m.p.	. 242°C
B088	Ħ	Ħ	coc4H9-t	3-01	4-CN	4-0CF ₃	0	m.p.	. 132°C
B089	æ	×	COC ₉ H ₁₉ -n	3-01	4-CN	4-0CF ₃	0	m.p.	. 163°C
B090	Ħ	Ħ	coch ₂ c1	3-C1	4-CN	4-0CF3	0	g.m.	. 191°C
B091	I	Ħ	CONH COL	3-01	4 CN	4-0CF3	0	m.p.	. 210°C
B092	Ħ	н	co ₂ cH ₃	3-C1	4-CN	4-0CF ₃	0		m.p. 199°C

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50	45		40	35	30	20	15	10		5	
					Table 5	(cont'd)		·			:
B093	H	H	н		3-C1	4-CN	4-0SO ₂ CF ₃		0	n.p.	m.p. 207°C
B094	<b>H</b>	Ħ	Ħ		3-C1	2-C1-4-CN	4-CF ₃	0	- <u>-</u>	n.p.	m.p. 203°C
B095	H	Ħ	н		3-C1	2-C1-4-CN	4-0CF3	0	<u> </u>	m.p.	196°C
B096	# 50	Ħ	H		3-C1	2-C1-4-CN	4-SCF3	0		m.p.	205°C
B097	1 Н	#	H		3-C1	2-CH ₃ -4-CN	4-C1		0	m.p.	183°C
B098	Н 8	Ħ	н		3-C1	2-CH ₃ -4-CN	4-0CF3	<u> </u>	0	m.p.	191°C
B099	н 6	Ħ	#		3-C1	2-CH ₃ -4-CN	4-SCF3	0	0	m.p.	192°C
   B100	Н 0	Ħ	H		3-C1	2-CH ₃ -4-CN	4-SOCF3		0	m.p.	180°C
B101	1 н	н	Ħ		3-C1	2-CH ₃ -4-CN	4-SO2CF3		<del>-</del>	m.p.	195°C
B102	2 H	H	Ħ		3-C1	3-CH3-4-CN	4-C1		<del>-</del>	m.p.	210°C
B103	3 H	×	н		3-C1	3-CH ₃ -4-CN	4-0CF3		<del>-</del>	m.p.	189°C
B104	4 H	н	ı		3-C1	3-CH ₃ -4-CN	4-SCF ₃		-	m.p.	179°C
B105	- H	#	н		3-01	3-CH ₃ -4-CN	4-SOCF3		<u> </u>	m.p.	188°C
B106	Н 9	<b></b>	Ħ		3-C1	3-CH3-4-CN	4-SO ₂ CF ₃		0	m.p.	191°C
B107	7 H	Ħ	Ħ		3-01	4-(CN) ₂	4-0CF ₃		0	m.p.	208°C

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5		m.p. 184°C	m.p. 172°C	m.p. 158°C	m.p. 185°C	m.p. 193°C	m.p. 183°C	m.p. 198°C	m.p. 200°C	m.p. 230°C	Glass-like amorphous substance	m.p. 131°C - isomer	m.p. 126°C + isomer	m.p. 191°C
	İ	0	0	0	0	0	0	0	0	0	0	0	0	0
10		4-CF ₃	4-0CF ₃	4-SCF ₃	4-0CF3	4-0CF ₃	4-C ₆ H ₁₃ -n	4-0CF ₃	4-0CF ₃	4-0CF ₃	4-0CF ₃	4-0CF ₃	4-ocr ₃	4-0CF ₃
		4,	4	4	4	4	4.	4	4	4	4,	4,	4	4,
20														
25	Table 5 (cont'd)	4-0CF ₃	4-0CHF ₂	4-0CHF ₂	$4-050_2$ CH ₃	$4-050_2$ CF $_3$	4-CN	4-CN	4-CN	4-CN	4-CN	4 -CN	4-CN	4-CN
	ည											<del></del>		
30	Table	3-C1	3-C1	3-C1	3-C1	3-C1	3-Br	3-Br	3-Br	3-Br	3-CHF ₂	3-CF3	3-CF3	3-CF3
35										ń				
40		<b></b>	#	<b></b>	Ħ	н	H	coch ₃	COC2H5	COC4H9-t	Ħ	Ħ	Ħ	сосн
		н	Ħ	н	н	H	H	Ħ	Ħ	H	Ħ	Ħ	Ħ	Ħ
45		н	Ħ	Ħ	Ħ	н	×	Ħ	Ħ	Ħ	н	н	ж	Н
5 <i>0</i>		B108	B109	B110	B111	B112	B113	B114	B115	B116	B117	B118	B119	B120

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5		. 208°C	. 183°C	. 251°C	. 190°C	Glass-like amorphous substance	Glass-like amorphous substance	. 189°C	. 153°C	). 146°C	). 166°C	p. 147°C	). 118°C
		m.p.	m.p.	m.p.	m.p.	Gla amo sub	Gla amo sub	m.p.	m.p.	m.p.	m.p.	m.p.	m.p.
		0	0	0	0	0	0	0	0	0	0	0	0
10		4-0CF ₃	4-0CF ₃	4-0CF ₃	4-OCF ₂ Br	4-SOCF ₃	4-so ₂ cF ₃	4-0CF ₃	4-CF ₃	4-0CF3	4-0CF3	4-0CF3	4-0CF ₃
75		4-	-4	-4	4	4	4,	4	4	कं	4	4	4
20	(cont'd)	4-CN	4-CN	4-CN	4-CN	4 -CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN	4-CN
25	Table 5 (cor	я 3	F3	સ ઝ	<del>й</del> 33	ਜ਼ 3	Έ3.	4-CF3	3-CH ₂ OCH ₃	3-CH ₂ OCH ₃	3-0CH ₃	3-0C3H7-i	3-0CHF ₂
30	Tak	3-CF3	3-CF3	OCF ₃ 3-CF ₃	3-CF3	3-CF3	3-CF ₃	4-	3–(	3–(	<u>3</u>		3–(
35		coc ₃ H ₇ -i	coc4H9-t	соин 🔘 о									
40		8	<del></del> -	<u>੪</u>	田	<b>#</b>	<b>=</b>	エ	<b>H</b>	_ <del>_</del>	<b></b>	<u> </u>	<b>=</b>
		H	Ħ	×	Ħ	H	Ħ	æ	Ħ	×	=	<b>H</b>	<b>#</b>
45		н	Ħ	Ħ	Ħ.	н	H	H	Ħ	Ħ	Ħ	H	=
50		B121	B122	B123	B124	B125	B126	B127	B128	B129	B130	B131	B132

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4-CN	4-CN	4 -CN	4-CN	3-0CHF ₂ 4-CN	H 3-0CHF ₂ 4-CN
3-0CF ₃ 4-CN	4-CN	4-CN	4-CN	H H 3-0CF ₃ 4-CN	H 3-0CF ₃ 4-CN
4-CN	4-CN	4-CN	4-CN	H H 3-0CF ₃ 4-CN	H 3-0CF ₃ 4-CN
4-CN	4-CN	4-CN	4-CN	H H 3-0CF ₃ 4-CN	H H H 3-0CF ₃ 4-CN
			3-0CF ₃	H H 3-0CF ₃	н н ж 3-0CF ₃
:0 ₂ СН ₃	202СН3	202СН3	3-0CH ₂ CO ₂ CH ₃	H H 3-0CH ₂ CO ₂ CH ₃	H H 3-0CH ₂ CO ₂ CH ₃
202СН3	202СН3	:0 ₂ СН ₃	3-0CF ₃ 3-0CF ₃ 3-0CF ₃ 3-0CF ₃	H H 3-0CF ₃ H H 3-0CF ₃ H H 3-0CF ₃ H H 3-0CF ₃	H       H       H       3-0CF3         H       H       H       3-0CF3         H       H       H       3-0CF3         H       H       H       3-0CF3
3-0CF 3-0CF 3-0CF	3-0CF 3-0CF 3-0CF	3-0CF 3-0CF 3-0CF		H H H H	H H H H H H
			<b># # # #</b>	* * * *	* * * * *

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Table 5 (cont'd)

B143	н	Н	н	4-F	4-NO ₂	.4-C1	လ	S m.p. 183°C
B144	Ħ	н	н	3-CF3	4-CH ₃	4-0CF3	0	O m.p. 166°C
B145	н	Ħ	COCO-OC2H5	3-c1	4-CN	4-0CF3	0	0 m.p. 192°C
B146	Ħ	Ħ	Ħ	н	3-CH=CH-CH=N-4	4-0CF3	w	S m.p. > 300°C Hydro-

Compounds B041 and B042 are diastereomers, and the Rf value of compound B041 is higher than that of compound B042. M.p. in the "physical properties" column

denotes melting point.

Table 6 shows 1-NMR data of the compounds having physical properties as glass like amorphous substance listed in Table 5.

# Table 6

10	No.	¹ H-NMR(CDC1 ₃ /TMS, & value, ppm.)
	B117	3.07(dd, lH), 3.18(dd, lH), 4.06-4.18(m, 2H),
		6.02(s, lH), 6.62(t, lH), 7.06-7.61(m, l2H),
15		7.73(s, lH).
	B125	3.12(dd, 1H), 3.23(dd, 1H), 4.12-4.32(m, 2H),
20		6.13(bs, 1H), 7.24-7.93(m, 12H), 8.08(bs, 1H).
	B126	3.11(dd, 1H), 3.23(dd, 1H), 4.13-4.28(m, 2H),
25		5.97(s, lH), 7.25-7.75(m, l2H),
20		7.90-8.00(bs, 1H).
	B134	3.05(dd, 1H), 3.18(dd, 1H), 4.05-4.15(m, 1H),
30		4.39(d, lH), 6.46(t, lH), 6.49(bs, lH),
		6.98-7.67(m, 12H), 8.04(s, 1H).
35	в138	3.04(dd, lH), 3.16(dd, lH), 4.03-4.21(m, 2H),
35		6.15(s, 1H), 6.95-7.65(m, 12H), 7.80(bs, 1H).
	B139	3.07(dd, 1H), 3.14(dd, 1H), 3.81(s, 3H),
40		3.97-4.11(m, 2H), 4.62(s, 2H), 5.72(s, 1H),
		6.79-7.63(m, 12H), 7.73(s, 1H).

- cont'd -

## Table 6 (cont'd)

As the compound of the general formula (VIII), i.e., the starting compound for producing the hydrazinecarboxamide derivative of the general formula (I) of the present invention, there may be used either commercially available one or one which is produced, for example, by the production process illustrated below.

wherein R3, R4, X and Y have the same meanings as those defined above, and R6 is a lower alkyl group.

That is, a compound of the general formula (VIII) can be produced by condensation reaction of a benzoic acid ester of the general formula (X) with a compound of the general formula (IX) in the presence of an inert solvent and a base.

The compound of the general formula (VI) can be produced, for example, by the production process illustrated below.

40 . 
$$W$$

$$N(R^1)-C-V + R^2-NH-NH_2$$
46 (XI) (VII)

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wherein R1, R2, Z and W have the same meanings as those defined above, and V is a halogen atom or a

leaving group such as a lower alkoxy group or an imidazole group.

That is, a compound of the general formula (VI) can be produced by reacting a compound of the general formula (XI) with a hydrazine derivative of the general formula (VII) in the presence of an inert solvent and a base.

Typical examples of the present invention are described below but should not be construed as limiting the scope of the invention.

#### Example 1

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1-1. Production of benzyl phenyl ketone hydrazone (compound No. V-1)

In 100 ml of ethanol was dissolved 5.0 g (26 mmoles) of benzyl phenyl ketone, followed by adding thereto 20 ml of hydrazine hydrate, and the reaction was carried out with stirring at a reaction temperature of 30° to 40° C for 6 hours.

After completion of the reaction, the solvent was removed by distillation under reduced pressure from the reaction solution containing the desired compound, and the residue was purified by a silica gel column chromatography (eluent, ethyl acetate: n-hexane = 1:4) to obtain 3.7 g of the desired compound as crystals.

Physical properties: m.p. 55°C. Yield: 69%.

1-2. Production of 2-(1,2-diphenylethylidene)-N-phenylhydrazinecarboxamide (compound No. A001)

In 30 ml of tetrahydrofuran was dissolved 0.50 g (2.4 mmoles) of benzyl phenyl ketone hydrazone, followed by adding thereto 0.5 ml of triethylamine and 0.28 g (2.4 mmoles) of phenyl isocyanate, and the reaction was carried out at room temperature for 3 hours.

After completion of the reaction, the reaction solution containing the desired compound was concentrated under reduced pressure, and the residue was purified by a silica gel column chromatography (eluent, chloroform) to obtain 0.47 g of the desired compound as crystals. Physical properties: m.p. 181 °C. Yield: 60%.

#### Example 2

5 Production of N-(4-chlorophenyl)-2-(1,2-diphenylethylidene)hydrazinecarboxamide (compound No. A004)

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In 30 ml of tetrahydrofuran was dissolved 0.62 g (3.0 mmoles) of benzyl phenyl ketone hydrazone, followed by adding thereto a drop of triethylamine and 0.45 g (3.0 mmoles) of 4-chlorophenyl isocyanate, and the reaction was carried out at room temperature for 3 hours.

After completion of the reaction, the solvent was removed by distillation under reduced pressure from the reaction solution containing the desired compound, and the crude crystals precipitated were washed with ether to obtain 0.90 g of the desired compound as crystals.

Physical properties: m.p. 199° C. Yield: 84%.

20 Example 3

3-1. Production of 4-fluorobenzylphenyl ketone hydrazone (compound No. V-2)

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In 50 ml of ethanol was dissolved 3.0 g (14 mmoles) of 4-fluorobenzylphenyl ketone, followed by adding thereto 3 ml of hydrazine hydrate and 10 mg of p-toluenesulfonic acid, and the reaction was carried out with heating under reflux for 2 hours.

After completion of the reaction, the excess hydrazine hydrate and the ethanol were distilled off under reduced pressure. Water was added to the residue and the desired compound was extracted with ether. The ether layer was washed with water and dried over anhydrous magnesium sulfate, after which the ether was removed to obtain 3.0 g of the desired compound as paste.

N-(4-chlorophenyl)-2-[2-(4-fluorophenyl)-1-phenylethylidene]hydrazinecarboxamide

Physical properties: paste. Yield: 93%.

3-2. Production o (compound No. A029)

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In 50 ml of tetrahydrofuran was dissolved 0.80 g (3.5 mmoles) of 4-fluorobenzylphenyl ketone hydrazone, followed by adding thereto 0.53 g (3.5 mmoles) of 4-chlorophenyl isocyanate, and the reaction was carried out at room temperature for 3 hours.

After completion of the reaction, the reaction solution containing the desired compound was concentrated under reduced pressure, and the residue was purified by a silica gel column chromatography (eluent, chloroform) to obtain 0.9 g of the desired compound as crystals. Physical properties: m.p. 213° C. Yield: 68%.

#### Example 4

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4-1. Production of 4-cyanobenzylphenyl ketone hydrazone (compound No. V-3)

In 200 ml of ethanol was dissolved 5.0 g (24 mmoles) of 4-cyanobenzylphenyl ketone, followed by adding thereto 15 ml of hydrazine hydrate and 15 mg of p-toluenesulfonic acid, and the reaction was carried out with heating under reflux for 4 hours.

After completion of the reaction, the excess hydrazine hydrate and the ethanol were distilled off under reduced pressure. Water was added to the residue and the desired compound was extracted with ether. The ether layer was washed with water and dried over anhydrous magnesium sulfate, after which the ether was removed to obtain 4.4 g of the desired compound as crystals.

Physical properties: m.p. 91 °C. Yield: 83%.

4-2. Production of 2-[2-(4-cyanophenyl)-1-phenylethylidene]-N-(4-trifluoromethylphenyl)-hydrazinecarboxamide (compound No. A050)

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In 30 ml of tetrahydrofuran was dissolved 1.0 g (4.4 mmoles) of 4-cyanobenzylphenyl ketone hydrazone, followed by adding thereto 0.81 g (4.0 mmoles) of 4-trifluoromethylphenyl isocyanate, and the reaction was carried out at room temperature for 2 hours.

After completion of the reaction, the reaction solution containing the desired compound was concentrated under reduced pressure, and the residue was purified by a silica gel column chromatography (eluent, chloroform) to obtain 0.85 g of the desired compound as crystals. Physical properties: m.p. 217 °C. Yield: 50%.

#### 25 Example 5

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5-1. Production of 4-nitrobenzylphenyl ketone hydrazone (compound No. V-4)

In 300 ml of ethanol was dissolved 5.0 g (21 mmoles) of 4-nitrobenzylphenyl ketone, followed by adding thereto 10 ml of hydrazine hydrate and 20 mg of p-toluenesulfonic acid, and the reaction was carried out at 60° C for 2 hours.

After completion of the reaction, the excess hydrazine hydrate and the ethanol were distilled off under reduced pressure. Water was added to the residue and the desired compound was extracted with ethyl acetate. The ethyl acetate layer was washed with water and dried over anhydrous magnesium sulfate, after which the ethyl acetate was removed to obtain 4.9 g of the desired compound as crystals. Physical properties: m.p. 72° C. Yield: 93%.

5-2. Production of 2-[2-(4-nitrophenyl)-1-phenyl-ethylidene)-N-(4-trifluoromethoxyphenyl)-hydrazinecarbothioamide (compound No. A080)

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In 30 ml of tetrahydrofuran was dissolved 0.80 g (3.1 mmoles) of 4-nitrobenzylphenyl ketone hydrazone, followed by adding thereto 0.62 g (2.8 mmoles) of 4-trifluoromethoxyphenyl isothiocyanate and three drops of triethylamine, and the reaction was carried out with heating under reflux for 5 hours. After completion of the reaction, the reaction solution containing the desired compound was concentrated under reduced pressure, and the residue was purified by a silica gel column chromatography (eluent, chloroform) to obtain 0.65 g of the desired compound as crystals.

Physical properties: m.p. 139°C. Yield: 49%.

#### 25 Example 6

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6-1. Production of 4-fluorobenzyl-4-fluorophenyl-ketone hydrazone (compound No. V-5)

$$F \longrightarrow \begin{array}{c} 0 \\ \parallel \\ C - CH_2 \longrightarrow \end{array} \longrightarrow \begin{array}{c} H_2N - N = C - CH_2 \longrightarrow \end{array} \longrightarrow F$$

In 50 ml of ethanol was dissolved 4.0 g (17 mmoles) of 4-fluorobenzyl-4-fluorophenylketone, followed by adding thereto 10 ml of hydrazine hydrate and a drop of concentrated sulfuric acid, and the reaction was carried out with heating under reflux for 2 hours.

After completion of the reaction the excess hydrazine hydrate and ethanol were distilled off under reduced pressure. Water was added to the residue and the desired compound was extracted with ether. The ether layer was washed with water and dried over anhydrous magnesium sulfate, after which the ether was removed under reduced pressure to obtain 4.2 g of the desired compound as paste. Physical properties: paste. Yield: 100%.

6.2. Production of 2-[1,2-bis(4-fluorophenyl)ethylidene]-N-(4-trifluoromethylphenyl)hydrazinecarboxamide (compound No. A227)

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In a mixture of 15 ml of pyridine and 15 ml of tetrahydrofuran was dissolved 0.50 g (2.0 mmoles) of 4-fluorobenzyl-4-fluorophenylketone hydrazone, followed by adding thereto 0.38 g (2.0 mmoles) of 4-trifluoromethylphenyl isocyanate, and the reaction was carried out at room temperature for 4 hours.

After completion of the reaction, the solvent was distilled off under reduced pressure and the residue was washed with an ether-n-hexane mixed solvent, whereby 0.55 g of the desired compound was obtained as crystals.

Physical properties: m.p. 199° C. Yield: 63%.

### Example 7

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Production of 2-(1,2-diphenylethylidene)-N-methyl-N-phenylhydrazinecarboxamide (compound No. A300)

In 30 ml of ethanol were dissolved 0.65 g (3.3 mmoles) of benzyl phenyl ketone and N-methyl-N-phenylhydrazinecarboxamide, followed by adding thereto a drop of concentrated sulfuric acid, and the reaction was carried out with heating under reflux for 8 hours. After completion of the reaction, the solvent was distilled off under reduced pressure, and the residue was purified by a silica gel column chromatography (eluent, ethyl acetate: n-hexane = 3:2) to obtain 0.26 g of the desired compound as crystals.

Physical properties: m.p. 113°C. Yield: 23%.

### Example 8

Production of 2-(1,2-diphenylethylidene)-1-methyl-N-phenylhydrazinecarboxamide (compound No. A303)

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In 50 ml of toluene was dissolved 1.0 g (5.1 mmoles) of benzyl phenyl ketone, followed by adding thereto 0.50 g (1.1 mmoles) of methylhydrazine and 0.20 g (1.2 mmoles) of p-toluenesulfonic acid, and the reaction was carried out with heating under reflux for 4 hours while eliminating water from the reaction system by azeotropy by means of a Dean-Stark trap.

After completion of the reaction, the solvent was distilled off under reduced pressure, and the oil thus obtained was dissolved in 20 ml of tetrahydrofuran, followed by adding thereto 0.58 g (4.9 mmoles) of phenyl isocyanate and 0.5 ml of triethylamine. The resulting mixture was allowed to stand overnight at room temperature to be subjected to reaction.

After completion of the reaction, and the residue was purified by a silica gel column chromatography (eluent, ethyl acetate: n-hexane = 1:5) to obtain 0.76 g of the desired compound as crystals.

Physical properties: m.p. 111° C. Yield: 46%.

### Example 9

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#### 9-1. Production of benzoin hydrazone (compound No. V-6)

In 50 ml of ethanol were dissolved 5.0 g (24 mmoles) of benzoin and 10 ml of hydrazine hydrate, followed by adding thereto a drop of concentrated sulfuric acid, and the reaction was carried out with heating under reflux for 2 hours.

After completion of the reaction, the excess hydrazine hydrate and ethanol were distilled off under reduced pressure. Water was added to the residue and the desired compound was extracted with ether. The ether layer was washed with water and dried over anhydrous magnesium sulfate, after which the ether was to obtain 5.1 g of the desired compound as paste. Physical properties: paste. Yield: 96%.

9-2. Production of 2-(2-hydroxy-1,2-diphenylethylidene)-N-phenylhydrazinecarboxamide (compound No. A330)

$$H_{2}N-N=C-CH \longrightarrow OH$$

$$NH-C-NH-N=C-CH \longrightarrow OH$$

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In 30 ml of tetrahydrofuran was dissolved 0.80 g (3.5 mmoles) of benzoin hydrazone, followed by adding thereto 0.42 g (3.5 mmoles) of phenyl isocyanate, and the reaction was carried out at room temperature for 4 hours.

After completion of the reaction, the solvent was distilled off under reduced pressure, and the residue was purified by a silica gel colume chromatography (eluent, tetrahydrofuran : chloroform = 1:10) to obtain 0.70 g of the desired compound as crystals.

Physical properties: m.p. 167° C. Yield: 57%.

#### Example 10 20

2-[2-(4-cyanophenyl)-1-phenylethyl]-N-(4-trifluoromethoxyphenyl)hydrazinecarboxamine Production of (compound No. B006)

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$$CF_{3}O - O - NH - C - NH - N = C - CH_{2} - CN$$

$$CF_{3}O - O - NH - C - NH - NHCHCH_{2} - CN$$

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#### 10-1.

In a mixture of 20 ml of tetrahydrofuran and 30 ml of methanol was dissolved 0.70 g (1.6 mmoles) of 2-[2-(4-cyanophenyl)-1-phenylethylidene]-N-(4-trifluorometh oxyphenyl)hydrazinecarboxamide, and 0.20 g (3.2 mmoles) of sodium cyanoborohydride was added to the solution. A saturated solution of hydrogen chloride in methanol was added dropwise with stirring at room temperature, and the reaction was carried out at room temperature for 1 hour.

After completion of the reaction, the solvent was distilled off under reduced pressure and ethyl acetate was added to the residue. The resulting mixture was neutralized with an aqueous sodium hydrogencarbonate solution and the ethyl acetate layer was separated. The thus obtained ethyl acetate containing the desired compound was dried over anhydrous magnedium sulfate, and the solvent was removed to obtain crude crystals of the desired compound.

The crude crystals obtained were washed with ether to obtain 0.54 g of the desired compound as crystals.

Physical properties: m.p. 204° C. Yield: 77%.

10-2.

In 80 ml of tetrahydrofuran was dissolved 1.0 g (2.3 mmoles) of 2-[2-(4-cyanophenyl)-1-phenylethylidene]-N-(4-trifluoromethoxyphenyl)hydrazinecarboxamide, and 0.1 g of 5% palladium carbon was added to the solution. The resulting mixture was allowed to absorb hydrogen gas at a pressure of 6 kg/cm² under shaking at room temperature for 60 hours.

After completion of the reaction, the catalyst was filtered off and the filtrate was evaporated under reduced pressure to obtain crude crystals of the desired compound.

The crude crystals obtained were washed with an ether-n-hexane mixture to obtain 0.97 g of the desired compound as crystals.

Physical properties: m.p. 204° C. Yield: 97%.

Example 11

Production of 2-[2-(4-nitrophenyl)-1-phenylethyl]-N-(4-trifluoromethoxyphenyl)hydrazinecarboxamide (compound No. B008)

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In a mixture of 10 ml of tetrahydrofuran and 10 ml of methanol was dissolved 0.48 g (1.0 mmole) of 2-[2-(4-nitrophenyl)-1-phenylethylidene]-N-(4-trifluorometh oxyphenyl)hydrazinecarboxamide, and 0.07 g (1 mmole) of sodium cyanoborohydride was added to the solution. Then, 5 ml of a saturated solution of hydrogen chloride in methanol was added dropwise with ice-cooling and the reaction was carried out for 30 minutes.

After completion of the reaction, the solvent was distilled off under reduced pressure and ethyl acetate was added to the residue. The resulting mixture was neutralized with an aqueous sodium hydrogencarbonate solution and the ethyl acetate layer was separated. The thus obtained ethyl acetate containing the desired compound was dried over anhydrous magnesium sulfate, and the solvent was removed to obtain crude crystals of the desired compound.

The crude crystals obtained were washed with ether-n-hexane to obtain 0.36 g of the desired compound as crystals.

Physical properties: m.p. 188° C. Yield: 75%.

50 Example 12

Production of 2-[1-(4-chlorophenyl)-2-(4-cyanophenyl)ethyl-N-(4-trifluoromethoxyphenyl)-hydrazinecarbothioamide (compound No. B033)

$$CF_{3}O \longrightarrow NH-C-NH-N=C-CH_{2} \longrightarrow CN$$

$$CF_{3}O \longrightarrow NH-C-NH-N+CHCH_{2} \longrightarrow C$$

In a mixture of 10 ml of tetrahydrofuran and 30 ml of methanol was dissolved 0.50 g (1.0 mmole) of 2-[1-(4-chlorophenyl)-2-(4-cyanophenyl)ethylidene]-N-(4-trifluoromethoxyphenyl)hydrazinecarbothioamide, and 0.19 g (3.1 mmoles) of sodium cyanoborohydride was added to the solution. Then, 5 ml of a saturated solution of hydrogen chloride in methanol was added dropwise at room temperature, and the reaction was carried out for 2 hours.

After completion of the reaction, the solvent was distilled off under reduced pressure and the ethyl acetate was added to the residue. The resulting mixture was neutralized with an aqueous sodium hydrogencarbonate solution and the ethyl acetate layer was separated. The thus obtained ethyl acetate containing the desired compound was dried over anhydrous magnesium sulfate, and the solvent was removed to obtain crude crystals of the desired compound.

The crude crystals obtained were washed with ether-n-hexane to obtain 0.27 g of the desired compound as crystals.

Physical properties: m.p. 195 °C. Yield: 54%.

#### Example 13

Production of 2-acetyl-2-[1-(3-chlorophenyl)-2-(4-cyanophenyl)ethyl]-N-(4-trifluoromethoxyphenyl)-hydrazinecarboxamide (compound No. B035)

$$CF_3O \longrightarrow NH-C-NH-NHCHCH_2 \longrightarrow CN$$

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In 30 ml of tetrahydrofuran was dissolved 0.70 g (1.5 mmoles) of 2-[1-(3-chlorophenyl)-2-(4-cyanophenyl)ethyl]-N-(4-trifluoromethoxyphenyl)hydrazinecarboxamide obtained in the same manner as in Example 10. To the resulting solution were added 0.35 g (4.4 mmoles) of acetyl chloride and 45 g (4.4 mmoles) of triethylamine at room temperature, and the reaction was carried out for 2 hours.

After completion of the reaction, the solvent was distilled off under reduced pressure, water was added to the residue and the desired compound was extracted with ethyl acetate. The ethyl acetate layer was washed with water and dried over anhydrous magnesium sulfate. The solvent was removed to obtain crude crystals of the desired compound.

The crude crystals obtained were washed with ether to obtain 0.55 g of the desired compound as crystals.

Physical properties: m.p. 178°C. Yield: 54%.

#### 25 Example 14

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Production of 2-[2-(4-cyanophenyl)-1-(3-trifluoromethylphenyl)ethylidene]-N-(4-trifluoromethoxy phenyl)-hydrazinecarboxamide (compound No. A261)

$$CF_3O \longrightarrow NH-C-NHN=C-CH_2 \longrightarrow CN$$
 $CF_3O \longrightarrow CF_3$ 

In 30 ml of tetrahydrofuran was dissolved 0.50 g (1.7 mmoles) of 4-cyanobenzyl-3-trifluoromethylphenylketone hydrazone, and 1 ml of pyridine was added. Then, a solution prepared by diluting 0.32 g (1.6 mmoles) of 4-trifluoromethoxyphenyl isocyanate with 5 ml of tetrahydrofuran was added dropwise with stirring at room temperature. After completion of the dropwise addition, the reaction was carried out at room temperature for another 4 hours.

After completion of the reaction, the solvent was distilled off under reduced pressure, and crude

crystals obtained was washed with ether-n-hexane to obtain 0.40 g of the desired compound as crystals. Physical properties: m.p. 191° C. Yield: 40%.

Insecticides containing the hydrazinecarboxamide derivative of the general formula (I) of the present invention as an active ingredient are suitable for controlling various insect pests such as agricultural insect pests, forest insect pests, horticultural insect pests, stored grain insect pests, sanitary insect pests, nematodes, etc. They have an insecticidal effect also, for example, on LEPIDOPTERA including summer fruit torrix (Adoxophyes orana fasciata), smaller tea tortrix (Adoxophyes sp.), Manchurian fruit moth (Grapholita inopinata), oriental fruit moth (Grapholita mlesta), soybean pod border (Leguminivora glycinivorella), mulberry leafroller (Olethreutes mori), tea leafroller (Caloptilla thevivora), Caloptilia sp. (Caloptilia zachrysa), apple leafminer (Phyllonorycter ringoniella), pear barkminer (Spulerina astaurota), common white (Piers rapae crucivora), tabacco budworm (Heliothis armigera), clodling moth (Laspeyresia pomonella), diamondback moth (Plutella xylostella), apple fruit moth (Argyresthia conjugella), peach fruit moth (Carposina niponensis), rice stem borer (Chilo suppressalia), rice leafroller (Cnaphalocrocis medinalis), tabacco moth (Ephestia elutella), mulberry pyralid (Glyphodes pyloalis), yellow rice borer (Scirpophaga incertulas), rice skipper (Parnara guttata), rice armyworm (Pseudaletia separata), pink borer (Sesamia inferens), common cutworm (Spodoptera litura), beet armyworm (Spodoptera exigua), etc.; HEMIPTERA including aster leafhopper (Macrosteles fascifrons), green rice leafhopper (Nephotettix cincticeps), brown rice planthopper (Nilaparvata lugens), whitebacked rice planthopper (Sogatella furcifera), citrus psylla (Diaphorina citri), grape whitefly (Aleurolobus taonabae), sweetpotato whitefly (Bemisia tabaci), greenhouse whitefly (Trialeurodes vaporariorum), turnip aphid (Lipaphis erysimi), green peach aphid (Myzus persicae), Indian wax scale (Ceroplastes ceriferus), cottony citrus scale (Pulvinaria aurantii), camphor scale (Pseudaonidia duplex), San Jose scale (Comstockaspis perniciosa), arrowhead scale (Unaspis yanonensis), etc.; COLEOPTERA including soybean beetle (Anomala rufocuprea), Japanese beetle (Popillia japonica), tobacco beetle (Lasioderma serricorne), powderpost beetle (Lyctus brunneus), twenty-eight-spotted ladybird (Epilachna vigintioctopunctata), adzuki bean weevil (Callosobruchus chinensis), vegetable weevil (Listroderes costirostris), maize weevil (Sitophilus zeamais), boll weevil (Anthonomus grandis grandis), rice water weevil (Lissorhoptrus oryzophilus), cucurbit leaf beetle (Aulacophora femoralis), rice leaf beetle (Outlema oryzae), striped flea beetle (Phyllotreta striolata), pine shoot beetle (Tomicus piniperda), Colorade potato beetle (Leptinotarsa decemilineata), Mexican bean beetle (Epilachna varivestis), corn rootworm (Diabrotica sp.), etc.: DIPTERA including melon fly (Dacus(Zeugodacus) cucurbitae), oriental fruit fly (Dacus-(Bactrocera) dorsalis), rice leafminer (Agromyza oryzae), onion maggot (Delia antiqua), seedcorn maggot (Delia platura), soybean pod gall midge (Asphodylia sp.), muscid fly (Musca domestica), house mosquite (Culex pipiens), etc.; and TYLENCHIDA including root-lesion nematode (Pratylenchus sp.), coffer root-lesion nematode (Pratylenchus coffeae), potato cyst nematode (Globodera rostochiensis), root-knot nematode (meloidogyne sp.), citrus nematode (Tylenchulus semipenetrans), Aphelenchus sp. (Aphelenchus avenae), chrysanthemum foliar nematode (Aphelenchoides ritzemabosi), etc. The insecticides are markedly effective particularly against insect pests belonging to LEPIDOPTERA, COLEOPTERA and the like.

The zoological names and the like are in accordance with Applied Zoology and Entomology Society of Japan, "List of Agricultural and Forest Injurious Animals and Insects", published in 1987.

The agricultural and horticultural insecticide of the present invention has a marked insecticidal effect on the above-exemplified insect pests, sanitary insect pests, and/or nematodes, which are injurious to paddy fields, fruit trees, vegetables and other crops, and flowers and ornament plants. Therefore, the desired effect of the insecticide of the present invention can be obtained by applying the insecticide to the paddy field water, stalks and leaves of fruit trees, vegetables, other crops, flowers and ornament plants, soil, etc., or to the inside of a house or ditches around a house, in which the above-exemplified sanitary insect pests injurious to men and beasts appear or are expected to appear. The application is carried out at a season at which the insect pests, sanitary insect pests or nematodes are expected to appear, before their appearance or at the time when their appearance is confirmed.

This invention however should not be limited to these embodiments.

When the hydrazinecarboxamide derivative of the general formula (I) of this invention is used as an insecticide, it is generally prepared into conveniently usable forms according to an ordinary manner for preparation of agrochemicals.

That is, the hydrazinecarboxamide derivative of the general formula (I) of this invention and, optionally, an adjuvant are blended with a suitable inert carrier in a proper proportion and prepared into a suitable preparation form such as a suspension, emulsifiable concentrate, soluble concentrate, wettable powder, granules, dust or tablet through dissolution, dispersion, suspension, mixing, impregnation, adsorption or sticking.

The inert carrier in this invention may be solid or liquid. Examples of the solid carrier are soybean flour,

cereal flour, wood flour, bark flour, saw dust, powdered tobacco stalks, powdered walnut shells, bran, powdered cellulose, extraction residues of vegetables, powdered synthetic polymers or resins, clays (e.g. kaolin, bentonite, and acid clay), talcs (e.g. talc and pyrophyllite), silica powders or flakes [e.g. diatomaceous earth, silica sand, mica and white carbon, i.e. synthetic, high-dispersion silicic acid, also called finely divided hydrated silica or hydrated silicic acid, some of commercially available products contain silicate as the major component)], activated carbon, powdered sulfur, powdered pumice, calcined diatomaceous earth, ground brick, fly ash, sand, calcium carbonate powder, calcium phosphate powder and other inorganic or mineral powders, chemical fertilizers (e.g. ammonium sulfate, ammonium phosphate, ammonium nitrate, urea and ammonium chloride), and compost. These carriers may be used alone or as a mixture thereof.

The liquid carrier is that which itself has solubility or which is without such solubility but is capable of dispersing an active ingredient with the aid of an adjuvant. The following are typical examples of the liquid carrier and can be used alone or as a mixture thereof. Water; alcohols such as methanol, ethanol, isopropanol, butanol and ethylene glycol; ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone, diisobutyl ketone and cyclohexanone; ethers such as ethyl ether, dioxane, Cellosolve, dipropyl ether and tetrahydrofuran; aliphatic hydrocarbons such as kerosene and mineral oils; aromatic hydrocarbons such as benzene, toluene, xylene, solvent naphtha and alkylnaphthalene; halogenated hydrocarbons such as dichloroethane, chloroform, carbon tetrachloride and chlorobenzene; esters such as ethyl acetate, diisopropyl phthalate, dibutyl phthalate and dioctyl phthalate; amides such as dimethylformamide, diethylformamide and dimethylacetamide; nitriles such as acetonitrile; and dimethyl sulfoxide.

The following are typical examples of the adjuvant, which are used depending upon purposes and used alone or in combination in some cases, or need not to be used at all.

To emulsify, disperse, dissolve and/or wet an active ingredient, a surfactant is used. Examples of the surfactant are polyoxyethylene alkyl ethers, polyoxyethylenealkylaryl ethers, polyoxyethylene higher fatty acid esters, polyoxyethylene resinates, polyoxyethylene sorbitan monoleate, polyoxyethylene sorbitan monoleate, alkylarylsulfonates, naphthalenesulfonic acid condensation products, ligninsulfonates and higher alcohol sulfate esters.

Further, to stabilize the dispersion of an active ingredient, tackify it and/or bind it, an adjuvant may be used. Examples of such an adjuvant are casein, gelatin, starch, methylcellulose, carboxymethylcellulose, gum arabic, polyvinyl alcohol, turpentine, bran oil, bentonite and ligninsulfonates.

To improve the flowability of a solid product, an adjuvant may be used. Examples of such an adjuvant are waxes, stearates and alkyl phosphates.

Adjuvants such as naphthalenesulfonic acid condensation products and polycondensates of phosphates may be used as a peptizer for dispersible products.

Adjuvants, e.g. silicon oils may be also used as a defoaming agent.

The content of the active ingredient may be varied as required. In dusts or granules, the suitable content thereof is from 0.01 to 50% by weight. In emulsifiable concentrates, flowable wettable powders, it is also from 0.01 to 50% by weight.

An insecticide containing the hydrazinecarboxamide derivative of the general formula (I) of this invention as an active ingredient is used to control a variety of insect pests in the following manner. That is, it is applied to the insect pests or a site where appearance of growth of the insect pests is undesirable, as it is or after being properly diluted with or suspended in water or the like, in an amount effective for control of the insect pests.

The amount of the insecticide containing the hydrazinecarboxamide derivative of the general formula (I) of this invention as an active ingredient is varied depending upon various factors such as a purpose, insect pests to be controlled, a growth state of a plant, tendency of insect pests appearance, weather, environmental conditions, a preparation form, an application method, an application site and an application time. It may be properly chosen in the range of 0.1 g to 5 kg (in terms of the active ingredient) per 10 ares depending upon purposes.

The insecticide containing the hydrozinecarboxamide derivative of the general formula (I) of this invention as an active ingredient may be used in admixture with other insecticides or fungicides in order to expand both spectrum of controllable insect pest species and the period of time when effective applications are possible or to reduce the dosage.

Typical preparation examples and test examples of the present invention are described below but should not be construed as limiting the scope of the invention.

In the preparation examples, parts are all by weight.

Formulation Example 1

Each compound of the invention 50 parts

Xylene 40 parts

Mixture of polyoxyethylene 10 parts

nonlyphenyl ether and calcium

alkylbenzenesulfonate

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An emulsifiable concentrate was prepared by mixing uniformly the above ingredients to effect dissolution.

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Formulation Example 2						
Each compound of the invention	3 parts					
Clay powder	82 parts					
Diatomaceous earth powder	15 parts					

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A dust was prepared by mixing uniformly and grinding the above ingredients.

# Formulation Example 3

Each compound of the invention	5 parts
Mixed powder of bentonite and clay	90 parts
Calcium lignin sulfonate	5 parts

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Granules were prepared by mixing the above ingredients uniformly, and kneading the resulting mixture together with a suitable amount of water, followed by granulation and drying.

### Formulation Example 4

	Each compound of the invention	20 parts
45	Mixture of kaolin and synthetic, high-dispersion silicic acid	75 parts
50	Mixture of polyoxyethylene nonylphenyl ether and calcium alkylbenzenesulfonate	5 parts

A wettable powder was prepared by mixing uniformly and grinding the above ingredients.

### Test Example 1

Insecticidal effect on common cutworm (Spodoptera litura)

A piece of cabbage leaf (cultivar: Shikidori) was immersed for about 30 seconds in a liquid chemical prepared by diluting a preparation containing each compound of this invention as an active ingredient to adjust the concentration to 500 ppm. After air-drying, it was placed in a plastic Petri dish having a diameter of 9 cm, and inoculated with second-instar larvae of common cutworm, after which the dish was closed and then allowed to stand in a room thermostated at 25° C. Eight days after the inoculation, the dead and alive were counted. The mortality was calculated according to the following equation and judgement was passed according to the criterion shown below. The test was carried out with triplicate groups of 10 insects.

	Criterion:						
	Degree of insecticidal effect	Mortality (%)					
Γ	Α	100					
	В	99 - 90					
-	С	89 - 80					
	D	79 - 50					
1	E	Less than 49					

The results obtained are shown in Table 7.

Table 7

5	Compound No.	Concent ration (ppm)	Judge- mwnr	Compound No.	Concent ration (ppm)	Judge- ment
10	A007	500	A	A059	500	A
	A016	500	С	A061	500	A
	A017	500	D	A062	500	A
15	A018	500	A	A063	500	A
	A019	500	С	A065	500	A
20	A020	500	A	A074	500	D
	A025	500	D	A076	500	A
	A027	500	A	A077	500	A
25	A028	500	A	A078	500	A
	A033	500	A	A079	500	A
	A034	500	A	A080	500	A
30	A038	500	A	A081	500	A
	A042	500	A	A082	500	A
35	A049	500	С	A087	500	A
	A050	500	С	A089	500	A
	A051	500	A	A090	500	A
40	A054	500	A	A091	500	A
	A055	500	A	A092	500	A
	A056	500	A	A093	500	A
45	A057	500	A	A096	500	A
	A058	500	A	A097	500	A
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Table 7 (cont'd)

5						
	A098	500	С	A135	500	A
	A099	500	A	A137	500	A
10	A101	500	A	A138	500	A
	A105	500	A	A140	500	A
1.5	A106	500	A	A141	500	A
15	A109	500	A	A142	500	A
į	Alll	500	A	A143	500	D
20	A112	500	С	A146	500	A
	A113	500	A	A147	500	A
	A114	500	A	A149	500	A
25	A115	500	A	A151	500	A
	A116	500	A	A152	500	A
<i>30</i>	A118	500	D	A153	500	A
	A120	500	מ	A155	500	A
	A125	500	A	A158	500	A
35	A127	500	A	A161	500	A
	A129	500	A	A164	500	. A
	A130	500	A	A165	500	A
40	A131	500	A	A167	500	A
	A132	500	A	A168	500	A
45	A133	500	A	A169	500	A
	A134	500	D	A170	500	Ä
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- cont'd -

Table 7 (cont'd)

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	A171	500	A	A218	500	A
	A172	500	A	A219	500	A
10	A173	500	A	A220	500	A
	A178	500	A	A221	500	A
4.5	A179	500	A	A225	500	A
15	A182	500	D	A226	500	A
	A185	500	A	A227	500	A
20	A189	500	A	A228	500	A
	A194	500	A	A229	500	A
	A195	500	A	A230	500	С
25	A196	500	A	A231	500	A
	A197	500	A	A232	500	A
	A198	500	A	A233	500	A
30	A199	500	A	A234	500	A
	A200	500	A	A235	500	A
35	A201	500	A	A239	500	A
	A202	500	A	- A240	500	A
	A203	500	A	A241	500	A
40	A212	500	С	A242	500	A
	A213	500	A	A248	500	A
	A214	500	С	A249	500	D
45	A216	500	A	A250	500	A
		1				

- cont'd -

Table 7 (cont'd)

5	A251	500	A	A321	500	A
	A254	500	D	A325	500	С
10	A257	500	A	A326	500	A
	A258	500	С	A342	500	D
	A260	500	A	A343	500	A
15	A261	500	A	A344	500	A
	A262	500	A	A354	500	D
20	A263	500	A	A362	500	A
20	A264	500	A	A367	500	A
	A265	500	A	A378	500	A
25	A269	500	A	A381	500	A
	A274	500	A	A383	500	A
i	A281	500	A	A385	500	A
30	A285	500	A	A386	500	A
	A292	500	С	88EA	500	С
25	A310	500	. <b>A</b>	A389	500	A
35	A312	500	A	A390	500	С
	A313	500	A	A393	500	A
40	A314	500	С	A394	500	С
	A318	500	D	A395	500	A
	A319	500	A	A397	500	· A
45	A320	500	A	A398	500	С

- cont'd -

Table 7 (cont'd)

5	A399	500	С	A427	500	A
	A400	500	A	A428	500	A
10	A401	500	A	A429	500	A
	A402	500	A	A430	500	D
	A403	500	A	A431	500	С
15	A404	500	A	A432	500	A
	A405	500	A	A433	500	A
	A406	500	A	A434	500	С
20	A409	500	. A	A436	500	A
	A410	500	A	A437	500	A
25	A411	500	A	A438	500	D
	A412	500	A	A439	500	A
	A415	500	A	A440	500	A
30	A418	500	A	A441	500	A
	A419	500	A	A442	500	A
05	A420	500	A	A443	500	A
35	A421	500	A	A447	500	A
	A422	500	A	A448	500	A
40	A423	500	A	A449	500	A
	A424	500	A	A450	500	A
	A425	500	A	A451	500	A
45	A426	500	С	A452	500	A
						<u> </u>

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Table 7 (cont'd)

5				1			}
į	A453	500	D	B007	500	A	
	A454	500	A	В008	500	A	
10	A456	500	D	B009	500	A	
	A460	500	A	B010	500	A	l
15	A461	500	A	B011	500	A	
	A462	500	A	B012	500	A	
	A463	500	A	B013	500	A	
20	A464	500	A	B014	500	A	
	A465	500	A	B015	500	A	
	A466	500	ם	B016	500	A	
25	A467	500	A	B018	500	A	
	A473	500	ם	B019	500	A	
30	A474	500	С	B020	500	A	
	A476	500	A	B021	500	A	١
	A477	500	A	B022	500	D	
35	A478	500	A	B023	500	A	
	A479	500	A	B024	500	A	
	A480	500	ם	B025	500	A	
40	B002	500	A	B026	500	A	۱
	B003	500	A	B027	500	D	
45	B004	500	A	B028	500	A	
40	в006	500	A	B030	500	A	١

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Table 7 (cont'd)

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5	B031	500	A	B061	500	D	
	B032	500	A	B062	500	A	
10	в033	500	A	в063	500	A	
	B034	500	A	В066	500	A	
	в035	500	A	в067	500	A	
15	в036	500	A	B068	500	A	
	B037	500	С	В069	500	A	۱
20	B043	500	A	B070	500	A	
20	B044	500	A	B071	500	A	
	B045	500	A	B072	500	A	
25	B047	500	D	B073	500	A	
	B048	500	A	B074	500	A	
	B049	500	A	в075	500	A	
30	в050	500	A	В079	500	A	
	B051	500	A	в080	500	A	
35	B052	500	A	во81	500	A	
33	в053	500	A	B082	500	A	
	B054	500	C,	B084	500	D	
40	B056	500	A	B085	500	A	
	B057	500	A	B086	500	A	
į	в059	500	D	B087	500	A	
45	B060	500	A	B088	500	A	
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- cont'd -

Table 7 (cont'd)

5	B090	500	A	B114	500	A
	B091	500	A	B115	500	A
10	B092	500	A	B116	500	A
	B093	500	A	B117	500	A
	B094	500	A	B118	500	A
15	B095	500	A	B119	500	A
	В096	500	A	B120	500	A
	B097	500	С	B121	500	A
20	в098	500	A	B122	500	A
	в099	500	A	B124	500	A
25	B100	500	A	B125	500	A
	B101	500	A	B126	500	A
	B103	500	A	B127	500	A
30	B104	500	A	B130	500	A
	в105	500	A	B131	500	D
	B106	500	D	B132	500	A
35	B107	500	A	B133	500	A
	B108	500	A	B134	500	A
40	В109	500	A	B135	500	A
	B110	500	A	B136	500	A
	B111	500	A	B137	500	A
45	B112	500	A	B138	500	A

- cont'd -

### Table 7 (cont'd)

1				1		
	B142	500	A	B145	500	A

Test Example 2

5 Insecticidal effect on adult maize weevil

### (Sitophilus zeamais)

Twenty to thirty grains of the brown rice were immersed for about 30 seconds in a liquid chemical prepared by diluting a preparation containing each compound of this invention as an active ingredient to adjust the concentration to 200 ppm. After air-drying, they were placed in a glass Petri dish having a diameter of 4 cm, and inoculated with adult maize weevils, after which the dish was closed and then allowed to stand in a room thermostated at 25° C. Eight days after the inoculation, the dead and alive were counted. The mortality was calculated according to the equation described in Test Example 1 and judgement was passed according to the criterion shown in Test Example 1. The test was carried out with triplicate groups of 10 insects.

The results obtained are shown in Table 8.

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Table 8

5	Compound No.	Concent ration (ppm)	Judge- mwnr	Compound No.	Concent ration (ppm)	Judge- ment
10	A004	200	D	A050	200	A
	A006	200	С	A056	200	A
	A007	200	Ā	A057	200	A
15	A015	200	A	A058	200	В
	A016	200	В	A059	200	A
i	A017	200	A	A061	200	D
20	A018	200	A	A062	200	D
	A020	200	A	A067	200	A
25	A021	200	A	A073	200	A
	A026	200	A	A077	200	A
	A027	200	A	A078	200	A
30	A028	200	A	A079	200	A
	A029	200	A	A080	200	D
	A030	200	A	A081	200	A
35	A033	200	A	A082	200	В
	A038	200	A	A091	200	A
40	A040	200	A	A092	200	A
<del>70</del>	A042	200	ם	A093	200	A
	A044	200	D	A096	200	A,
45	A046	200	С	A097	200	A

- cont'd -

Table 8 (cont'd)

5 _						
	A098	200	A	A135	200	A
	A099	200	A	A137	200	A
10	A101	200	A	A138	200	A
	A102	200	D	A140	200	A
15	A103	200	A	A141	200	D
15	A105	200	A	A146	200	A
	A106	200	A	A147	200	A
20	A110	200	A	A148	200	A
	A111	200	A	A149	200	A
	A112	200	A	A150	200	В
25	A113	200	A	A151	200	A
	A114	200	A	A152	200	D
30	A115	200	С	A154	200	A
30	A116	200	A	A155	200	A
	A118	200	A	A156	200	A
35	A123	200	В	A163	200	С
	A125	200	A	A164	200	A
	A127	200	A	A165	200	A
40	A131	200	A	A161	200	D
	A132	200	A	A162	200	D
45	. A133	200	A	A166	200	С
45	A134	200	A	A167	200	В
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- cont'd -

Table 8 (cont'd)

5				78	·		
	A168	200	A	A203	200	A	l
	A170	200	A	A204	200	A	l
10	A172	200	D	A212	200	מ	
	A173	200	С	A213	200	D	
15	A175	200	D	A215	200	A	
70	A178	200	A	A216	200	A	
	A179	200	В	A217	200	A	
20	A184	200	A	A218	200	A	l
	A185	200	A	A219	200	A	
	A186	200	С	A220	200	A	
25	A187	200	С	A221	200	A	
	A188	200	D	A223	200	A	ļ
30	A189	200	A	A224	200	A	
30	A194	200	A	A225	200	A	١
	A195	200	A	A226	200	A	
35	A196	200	A	A227	200	A	
	A197	200	A	A228	200	A	
	A198	200	A	A229	200	A	
40	A199	200	A	A230	200	В	
·	A200	200	A	A231	200	A	
45	A201	200	A	A232	200	A	
40	A202	200	A	A233	200	D	
							i

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Table 8 (cont'd)

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	A234	200	A	A273	200	D
	A235	200	A	A283	200	A
10	A236	200	D	A284	200	A
	A237	200	A	A285	200	A
	A238	200	A	A288	200	A
15	A239	200	A	A289	200	С
	A240	200	A	A292	200	A
20	A241	200	A	A293	200	D
	A242	200	A	A294	200	В
	A257	200	A	A295	200	С
25	A258	200	С	A319	200	A
	A259	200	A	A320	200	A
	A260	200	A	A321	200	A
30	A261	200	A	A326	200	D
	A262	200	A	A351	200	A
35	A263	200	A	A370	200	A
	A264	200	A	A374	200	ā
	A265	200	A	A380	200	С
40	A268	200	A	A381	200	A
	A269	200	A	A383	200	A
	A270	200	A	A385	200	A
45	A272	200	D	A386	200	A
		Į				

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Table 8 (cont'd)

5	A388	200	С	A421	200	A
	A389	200	A	A422	200	A
10	A390	200	С	A423	200	A
	A393	200	A	A424	200	D
	A394	200	С	A425	200	A
15	A395	200	A	A427	200	A
	A397	200	A	A429	200	D
20	A398	200	С	A430	200	В
20	A400	200	A	A436	200	A
	A401	200	A	A439	200	A
25	A402	200	A	A440	200	A
	A403	200	A	A441	200	A
	A404	200	A	A442	200	A
30	A405	200	В	A443	200	A
	A406	200	D	A447	200	A
35	A409	200	A	A448	200	A
	A410	200	В	A449	200	A
	A411	200	A	A450	200	A
40	A415	200	A	A451	200	A
	A418	200	A	A452	200	A
	A419	200	A	A453	200	D 1.
45	A420	200	A	A454	200	A

- cont'd -

Table 8 (cont'd)

5	A460	200	A	B018	200	A
	A461	200	A	B019	200	A
10	A462	200	A	В020	200	A
	A463	200	В	B021	200	С
	A464	200	A	B022	200	D
15	A465	200	A	B023	200	A
	A466	200	A	B024	200	A
	A467	200	A	B025	200	A
20	в001	200	A	B026	200	A
	B002	200	A	B028	200	В
25	в003	200	A	B031	200	A
	B004	200	A	B032	200	A
	воо6	200	A	B034	200	A
30	воов	200	A	в035	200	A
	воо9	200	A	в036	200	A
	в010	200	A	в037	200	A
35	B011	200	A	B044	200	A
	в012	200	A	B045	200	A
40	B013	200	A	B047	200	A
	B014	200	A	B048	200	A
	в015	200	A	B049	200	D
45	B016	200	A	в050	200	A
	}					

- cont'd -

Table 8 (cont'd)

5		·	<u> </u>		1	
	B051	200	A	B082	200	A
	B052	200	D	B084	200	A
10	в055	200	A	B085	200	A
	B056	200	Α .	B086	200	В
10	B057	200	A	B087	200	A
15	B060	200	A	В088	200	В
	B061	200	A	B090	200	A
20	B062	200	A	B091	200	В
	B063	200	С	B092	200	A
	B066	200	A	B093	200	A
25	B067	200	c ·	B094	200	A
	B068	200	A	В095	200	A
	B069	200	A	в096	200	A
30	B070	200	A	B097	200	С
	B071	200	A	в098	200	В
35	B072	200	A	B107	200	A
	B073	200	A	B108	200	A
	B074	200	A	B109	200	A
40	В075	200	A	B110	200	A
	В079	200	A	B111	200	A
	B080	200	A	B112	200	A
45	B081	200	A	B114	200	A

- cont'd -

### Table 8 (cont'd)

5			r			
	B115	200	A	B126	200	A
	B116	200	A	B132	200	A
10	B117	200	A	В133	200	A
	B118	200	A	B134	200	A
	B119	200	A	в135	200	A
15	B120	200	A	B136	200	A
	B121	200	A	B137	200	A
20	B122	200	A	в138	200	A
	B124	200	A	B145	200	A
	в125	200	A			
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#### Claims

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1. A hydrazinecarboxamide derivative represented by the general formula (I):

$$\begin{array}{c}
Z \\
\downarrow \\
N(R^1)-C-N(R^2)-A-C \\
\downarrow \\
R^3
\end{array}$$
(1)

wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkoxy group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R⁴ is a hydrogen atom or a lower alkyl group, R³ and R⁴ being able to be taken together to represent an oxygen atom, A is

$$-N=C(-\sqrt{X})-or-N(R^5)-CH(-\sqrt{X})-$$

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(wherein X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkoxyalkyl groups; lower alkenyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower a ycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, and R5 is a hydrogen atom; a lower alkylcarbonyl group; a lower haloalkylcarbonyl group; a cycloalkylcarbonyl group; a lower alkoxycarbonyl group; a lower alkoxydicarbonyl group; an unsubstituted phenylcarbonyl group; substituted phenylcarbonyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; or a substituted aminocarbonyl group having 1 or 2 substituents which may be the same or different and are selected from the group consisting of hydrogen atom. lower alkyl groups, unsubstituted phenyl group, and substituted phenyl groups having 1 to 5 substituents which may be the same or different and are selected from halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups), Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsulfonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylsulfonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atoms, halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom.

2. A hydrazinecarboxamide derivative according to Claim 1, wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom or a lower alkyl group, R⁴ is a hydrogen atom or a lower alkyl group, A is

$$-N=C(-\sqrt{X})- \text{ or } -N(R^5)-CH(-\sqrt{X})-$$

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(wherein X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom, halogen atoms, lower alkyl groups and lower haloalkyl groups, and R⁵ is a hydrogen atom), Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom, halogen atoms, cyano group and nitro group, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom, halogen atoms, lower haloalkyl groups, lower haloalkoxy groups, lower alkylthio groups, lower haloalkylsulfinyl groups, lower haloalkylsulfinyl groups, lower alkylsulfonyl groups, and W is an oxygen atom.

- 3. A hydrazinecarboxamide derivative according to Claim 2, wherein X is at the 3-position and Y or Z has a substituent at the 4-position.
- 4. A process for producing a hydrazinecarboxamide derivative represented by the general formula (I'):

$$\begin{array}{c|c}
 & X & Y \\
 & X & X & Y \\
 & X & X & Y \\
 & X & X & X & Y \\
 & X & X & X & Y \\
 & X & X & X & Y \\
 & X & X & X & Y \\
 & X & X & X & Y \\
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 & X & X & X & X & X & X & X & X \\
 & X & X & X & X & X & X &$$

(wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkoxy group; a lower

alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R4 is a hydrogen atom or a lower alkyl group, R3 and R4 being able to be taken together to represent an oxygen atom, X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkoxyalkyl groups; lower alkoxycarbonyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsulfonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and

are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group

consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower haloalkylsulfinyl groups; lower haloalkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower alkylsulfonyl groups; lower alkylsulfonyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom) which comprises reacting a compound represented by the general formula (VIII):

(wherein  $R^3$ ,  $R^4$ , X and Y have the same meanings as those defined above) with a compound represented by the general formula (VI):

$$\begin{array}{c} Z \\ W \\ \parallel \\ -\text{N}(R^1) - \text{C} - \text{N}(R^2) - \text{NH} \end{array}$$
 (VI)

(wherein R1, R2, Z and W have the same meanings as those defined above).

5. A process for producing a hydrazinecarboxamide derivative represented by the general formula (I'):

$$\begin{array}{c|c}
 & X \\
 & X \\
 & X \\
\end{array}$$

$$\begin{array}{c|c}
 &$$

(wherein R¹ is a hydrogen atom, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R⁴ is a hydrogen atom or a lower alkyl group, R³ and R⁴ being able to be taken together to represent an oxygen atom, X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl

groups; lower alkoxy groups; lower haloalkoxy groups; lower alkoxyalkyl groups; lower alkenyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsulfonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylcarbonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom) which comprises reacting a compound represented by the general formula (VIII):

$$\begin{array}{c|c}
X & 0 & R^3 & Y \\
 & \parallel & \parallel & \\
 & -C - C & \\
 & - Q & 4
\end{array}$$
(VIII)

(wherein R³, R⁴, X and Y have the same meanings as those defined above) with a hydrazine derivative represented by the general formula (VII):

### R²-NHNH₂ (VII)

(wherein R² has the same meaning as that defined above) to obtain a compound represented by the general formula (V):

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$$NH(R^{2})-N=C - \begin{matrix} R^{3} & Y \\ C & \\ R^{4} \end{matrix}$$

$$(V)$$

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(wherein  $R^2$ ,  $R^3$ ,  $R^4$ , X and Y have the same meanings as those defined above), and reacting this compound with a compound represented by the general formula (IV):

$$Z$$
 $N=C=W$  (IV)

(wherein Z and W have the same meanings as those defined above) after or without isolating the same.

6. A process for producing a hydrazinecarboxamide derivative represented by the general formula (I"):

(wherein R1 is a hydrogen atom or a lower alkyl group, R2 is a hydrogen atom or a lower alkyl group, R3 is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkoxy group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R4 is a hydrogen atom or a lower alkyl group, R3 and R4 being able to be taken together to represent an oxygen atom, X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkoxyalkyl groups; lower alkenyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfonyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxy carbonyl groups; unsubstituted amino group; substituted amino

groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower akylsulfonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylcarbonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom) which comprises subjecting a compound represented by the general formula (I'):

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$$\begin{array}{c|c}
 & X & Y \\
 & X & X & Y \\
 & X & X & X \\
\end{array}$$

$$\begin{array}{c|c}
 & X & Y \\
 &$$

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(wherein R¹, R², R³, R⁴, X, Y, Z and W have the same meanings as those defined above) to reduction reaction in the presence of a reducing agent.

## 7. A process for producing a hydrazinecarboxamide derivative represented by the general formula (I"'):

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(wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R⁴ is a hydrogen atom or a lower alkyl group, R³ and R⁴ being able to be taken together to represent an oxygen atom, X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group;

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lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkoxyalkyl groups; lower alkenyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, and R5 is a hydrogen atom; a lower alkylcarbonyl group; a lower haloalkylcarbonyl group; a cycloalkylcarbonyl group; a lower alkoxycarbonyl group; a lower alkoxydicarbonyl group; an unsubstituted phenylcarbonyl group; substituted phenylcarbonyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; or a substituted aminocarbonyl group having 1 or 2 substituents which may be the same or different and are selected from the group consisting of hydrogen atom, lower alkyl groups, unsubstituted phenyl group, and substituted phenyl groups having 1 to 5 substituents which may be the same or different and are selected from halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsufonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylcarbonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyl groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom) which comprises reacting a hydrazinecarboxamide derivative represented by the general formula (I"):

(wherein R¹, R², R³, R⁴, X, Y, Z and W have the same meanings as those defined above) with a halide represented by the general formula (III):

(wherein R^{5'} is a lower alkylcarbonyl group; a lower haloalkylcarbonyl group; a cycloalkylcarbonyl group; a lower alkoxycarbonyl group; a lower alkoxydicarbonyl group; an unsubstituted phenylcarbonyl group; a substituted phenylcarbonyl group having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; or a substituted aminocarbonyl group having 1 to 2 substituents which may be the same or different and are selected from the group consisting of hydrogen atom, lower alkyl groups, unsubstituted phenyl group, and substituted phenyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and Hal is a halogen atom) or an isocyanate represented by the general formula (II):

(wherein R^{5"} is a lower alkyl group; an unsubstituted phenyl group; or a substituted phenyl group having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups).

8. An agricultural and horticultural insecticide comprising as an active ingredient a hydrazinecarboxamide derivative represented by the general formula (I):

wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkoxy group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R⁴ is a hydrogen atom or a lower alkyl group, R³ and R⁴ being able to be taken together to represent an oxygen atom, A is

$$-N=C(-\sqrt{X})-or-N(R^5)-CH(-\sqrt{X})-$$

(wherein X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkyl groups; lower alkoxyalkyl groups; lower alkenyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups

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which may be the same or different; and dioxolane group, and R5 is a hydrogen atom; a lower alkylcarbonyl group; a lower haloalkylcarbonyl group; a cycloalkylcarbonyl group; a lower alkoxycarbonyl group; a lower alkoxydicarbonyl group; an unsubstituted phenylcarbonyl group; substituted phenylcarbonyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; or a substituted aminocarbonyl group having 1 or 2 substituents which may be the same or different and are selected from the group consisting of hydrogen atom, lower alkyl groups, unsubstituted phenyl group, and substituted phenyl groups having 1 to 5 substituents which may be the same or different and are selected from halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups), Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloakoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsulfonyl groups, and substituted aminocarbonyl groups having as the substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; tower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylcarbonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower akkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom.

40 9. An agricultural and horticultural insecticide according to Claim 8, wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom or a lower alkyl group, A is

$$-N=C(-V)$$
 )- or  $-N(R^5)-CH(-V)$  )-

(wherein X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom, halogen atoms, lower alkyl groups and lower haloalkyl groups, and R⁵ is a hydrogen atom), Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom, halogen atoms, cyano group and nitro group, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom, halogen atoms, lower haloalkyl groups, lower haloalkoxy groups, lower alkylthio groups, lower haloalkylthio groups, lower haloalkylsulfinyl groups, lower alkylsulfonyl groups, and W is an oxygen atom.

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- 10. An agricultural and horticultural insecticide according to Claim 9, wherein X is at the 3-position and Y or Z has a substituent at the 4-position.
- 11. A process for controlling undesirable insect pests which comprises applying an agricultural and horticultural insecticide comprising as an active ingredient a hydrazinecarboxamide derivative represented by the general formula (I):

$$\begin{array}{c|c}
Z \\
 & W \\
 & \parallel \\
 &$$

wherein R¹ is a hydrogen atom or a lower alkyl group, R² is a hydrogen atom or a lower alkyl group, R³ is a hydrogen atom; a hydroxyl group; a lower alkyl group; a lower alkoxy group; a lower alkylcarbonyloxy group; an unsubstituted phenylcarbonyloxy group; or a substituted phenylcarbonyloxy group having on the phenyl ring 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups and lower haloalkyl groups, R⁴ is a hydrogen atom or a lower alkyl group, R³ and R⁴ being able to be taken together to represent an oxygen atom, A is

$$-N=C(-\sqrt{X})-or -N(R^5)-CH(-\sqrt{X})-$$

(wherein X represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; lower alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkoxyalkyl groups; lower alkenyloxy groups; cycloalkylcarbonyloxy groups; lower alkoxycarbonyloxy groups; lower alkoxycarbonylalkyloxy groups; lower alkylcarbonylalkyloxy groups; lower alkylsulfonyloxy groups; phenoxy group; methylenedioxy group; alkenylene groups having 3 to 4 carbon atoms so as to form a polycyclic ring together with the adjacent carbon atom of the phenyl ring; unsubstituted amino group; substituted amino groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminocarbonyloxy groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; and dioxolane group, and R5 is a hydrogen atom; a lower alkylcarbonyl group; a lower haloalkylcarbonyl group; a cycloalkylcarbonyl group; a lower alkoxycarbonyl group; a lower alkoxydicarbonyl group; an unsubstituted phenylcarbonyl group; substituted phenylcarbonyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; or a substituted aminocarbonyl group having 1 to 2 substituents which may be the same or different and are selected from the group consisting of hydrogen atom, lower alkyl groups, unsubstituted phenyl group, and substituted phenyl groups having 1 to 5 substituents which may be the same or different and are selected from halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups), Y represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; hydroxyl group; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkenyloxy groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkoxycarbonyl groups; unsubstituted amino group; substituted amino groups having 1 or 2 substituents selected from the group consisting of formyl group, lower alkylcarbonyl groups, lower alkylsulfonyl groups, and substituted aminocarbonyl groups having as the

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substituent(s) one or more lower alkyl groups which may be the same or different; unsubstituted aminocarbonyl group; substituted aminocarbonyl groups having as the substituent(s) 1 or 2 lower alkyl groups which may be the same or different; substituted aminosulfonyl groups having as the substituent-(s) 1 or 2 lower alkyl groups which may be the same or different; phenyl group; or azaalkenylene groups having 2 to 3 carbon atoms so as to form a polycyclic ring together with the adjacent atom of the phenyl group, Z represents 1 to 5 atoms or groups which may be the same or different and are selected from the group consisting of hydrogen atom; halogen atoms; cyano group; nitro group; alkyl groups; lower haloalkyl groups; unsubstituted cycloalkyl groups; substituted cycloalkyl groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms and lower alkyl groups; lower alkoxy groups; lower haloalkoxy groups; lower alkylthio groups; lower haloalkylthio groups; lower alkylsulfinyl groups; lower haloalkylsulfinyl groups; lower alkylsulfonyl groups; lower haloalkylsulfonyl groups; lower alkylcarbonyl groups; lower alkoxycarbonyl groups; lower alkylcarbonyloxy groups; lower alkylsulfonyloxy groups; lower haloalkylsulfonyloxy groups; unsubstituted phenoxy group; substituted phenoxy groups having 1 to 5 substituents which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups; unsubstituted pyridyloxy group; substituted pyridyloxy groups having as the substituent(s) 1 to 4 atoms or groups which may be the same or different and are selected from the group consisting of halogen atoms, lower alkyl groups, lower haloalkyl groups, lower alkoxy groups and lower haloalkoxy groups, and W is an oxygen atom or a sulfur atom) in an amount of 0.1 g to 5 kg in terms of the active ingredient per 10 ares in order to protect useful crops against undesirable insect pests.



## EUROPEAN SEARCH REPORT

Application Number

EP 91 10 9275

D	OCUMENTS CONS				
Category		th indication, where appropriate, evant passages		lelevant to claim	CLASSIFICATION OF THE APPLICATION (Int. CI.5)
×	page 805, abstract no. 1847 EL-REASH et al.: "Synthesi	ol. 110, no. 20, 15 May 198 780y, Columbus, Ohio, US; is and characterization of fiverived from benzoin thiosem	А. ⁄ө		C 07 C 281/06 C 07 C 281/14 C 07 C 335/40 A 01 N 47/34
x	September 1976, page 632 Ohio, US; A.B. TOMCHIN e	OF JAPAN vol. 85, no. 13, 2: , abstract no. 94317p, Colur t al.: "Semicarbazones and acyclic and carbocyclic seri acety!"	nbus,		
Α	EP-A-0 144 853 (FUJISAV * claim 1 *	VA PHARMACEUTICAL)	1		
Α	DE-B-2 304 789 (PHILIPS) * claim 1 *	) 	1		
Α	GB-A-1 314 899 (STERLIN * claim 14 *	NG-WINTHROP)	1		TECHNICAL FIELDS
Α	PATENT ABSTRACTS OF November 1979; & JP - A - 54119029 (MEIJI	SEIKA) 14.09.1979	4), 16 1		SEARCHED (Int. CI.5)  C 07 C 281/06 C 07 C 281/14 C 07 C 335/40 A 01 N 47/34
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